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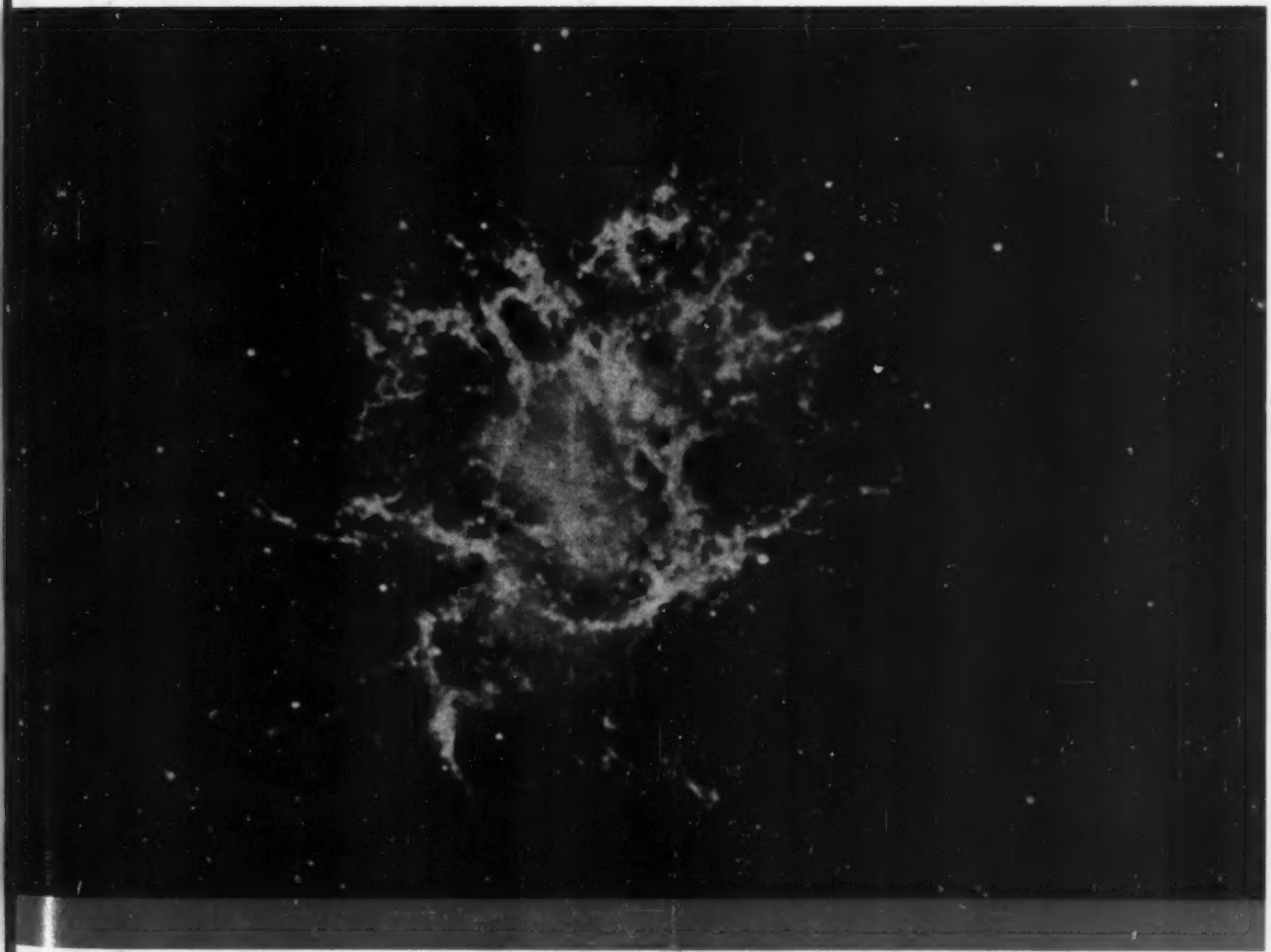

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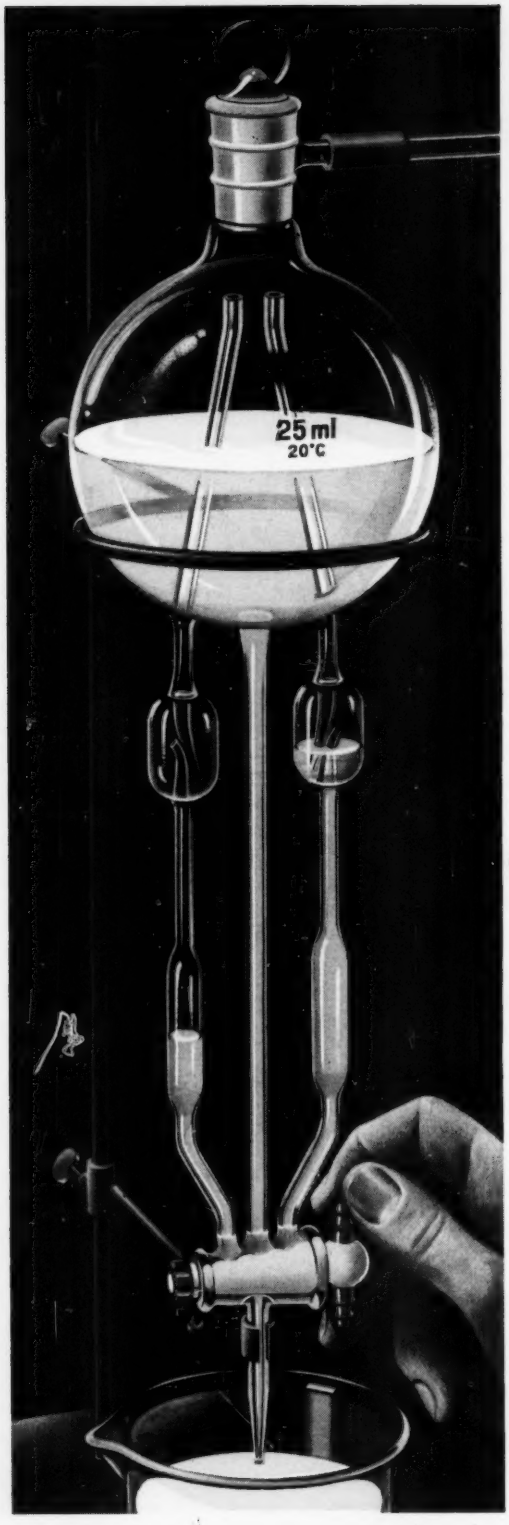

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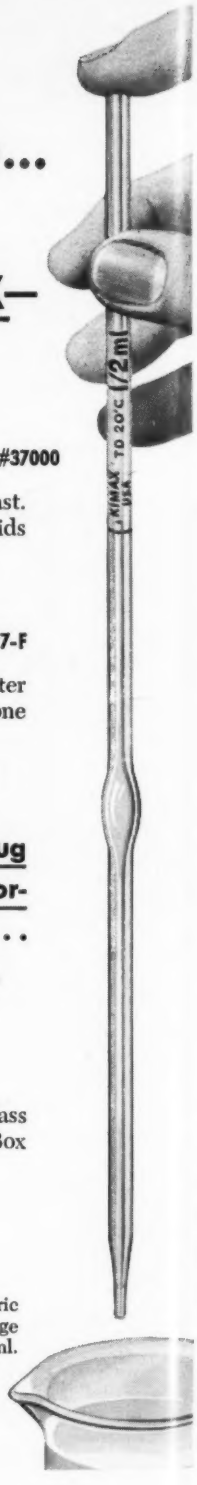

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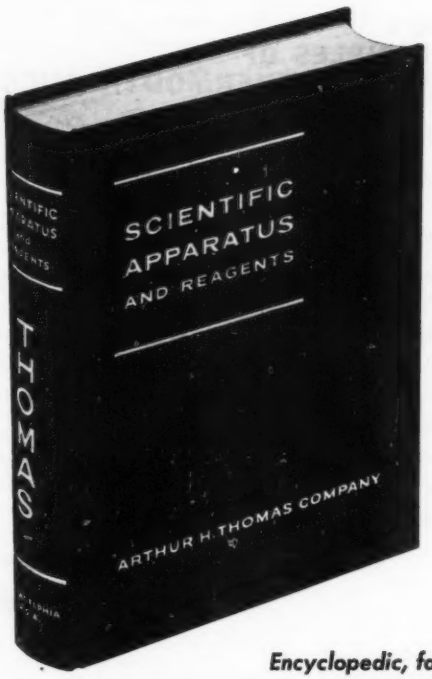

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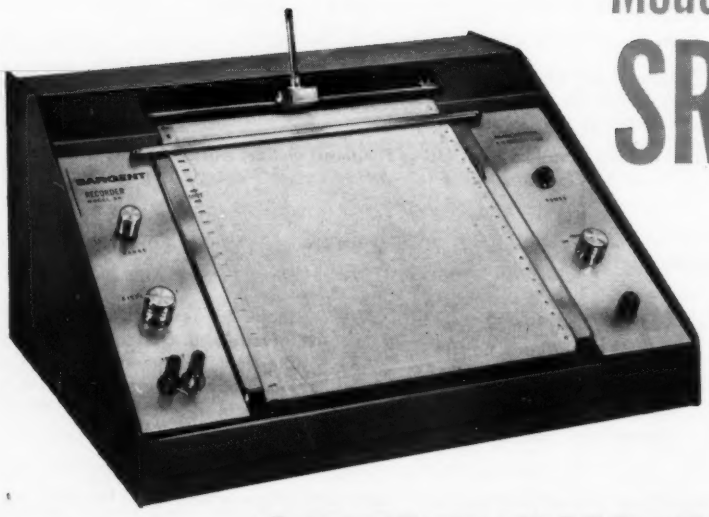

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This series of articles presents a survey of the basic principles, characteristics, and limitations of those instruments which find important applications in chemical work. The emphasis is on commercially available equipment, and approximate prices are quoted to show the order of magnitude of cost of the various types of design and construction.

11. Infrared Spectrometers (Concluded)

Applied Physics Corporation

One of the most highly developed infrared spectrometers commercially available is the recently-announced Cary-White Model 90 instrument manufactured by Applied Physics Corporation, Monrovia, California. This is a double-beam, prism-grating double monochromator, ratio-recording spectrometer, as indicated by the optical diagram shown in Figure 26.

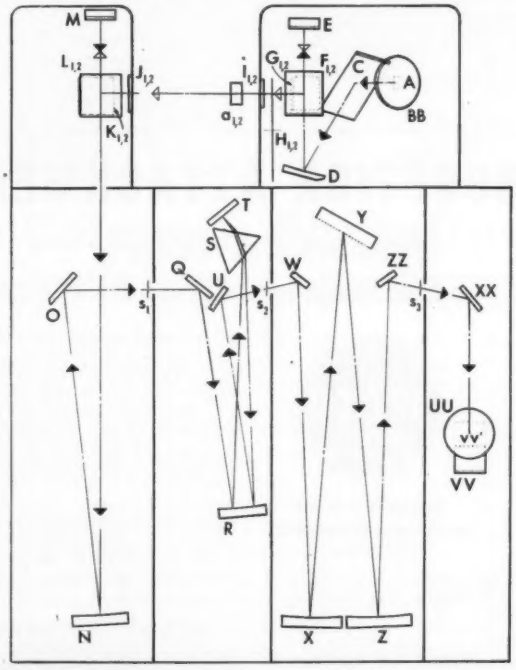

Figure 26. Optical diagram of the Cary-White Model 90 Infrared Spectrometer.

Radiation from the source, A, which may be either a Nernst glower or a nichrome ribbon, is directed by the parabolic condensing mirror, BB, behind it onto the flat mirror, C, which reflects it to the auxiliary condenser, D. From here it goes to the multifaceted beam splitter, E, which consists of a stack of alternately-oriented reflectors (*cf.*, Figure 34 of section on Spectrophotometers, September, 1960). The beam splitter divides the beam into two beams directed downward toward the mirrors F_1 and F_2, which reflect the beams horizontally to the diagonal mirrors, G_1 and G_2. These redirect the beams through the two chopper discs, H_1 and H_2, which are slotted so as to chop the sample beam $13^1/_3$ times per second, and the reference beam $26^2/_3$ times per second. These chopped beams leave the source compartment through the windows I_1 and I_2 (one below the other), and pass through the sample and reference cells, a_1 and a_2. They enter the beam combiner compartment through windows J_1 and J_2 and fall on the diagonal mirrors K_1 and K_2. These send the beams to another pair of mirrors, L_1 and L_2, which send them to the beam combiner, M, a multifaceted stack similar to the original beam splitter, E. The combined beam is directed to spherical mirror, N, to another spherical mirror, O, and is focussed onto the entrance slit, s_2 of the monochromator.

The energy from the entrance slit falls upon the aberration compensator, Q, and thence to the collimating mirror, R, which sends it to the KBr prism, S, backed by the Littrow mirror, T. After being dispersed twice by its entrance and return path through the prism, the radiation is reflected by the collimator, R, to the aberration compensator, U, and thence to the intermediate slit, s_2, through which it enters the grating monochromator. The aberration compensator, W, sends the beam to the collimating mirror, X, which directs a parallel beam onto the grating, Y. From the grating, the dispersed radiation falls on the second collimator, Z, which sends it to an aberration compensator, ZZ, that directs it to the exit slit, s_3. In the detector section, the toroidal focusing mirror, XX, sends the radiation to either a mirror, VV, or a LiF filter, VV' (to eliminate stray radiation) which reflects it to the ellipsoidal mirror, UU, that focuses the beam onto the detector.

The amplified output from the detector consists of two frequencies; a $13^1/_3$ cps alternation corresponding to the intensity of the beam which passed through the sample channel, and a $26^2/_3$ cps alternation corresponding to the reference beam. These two signals are sorted out electronically, and their ratio is presented to the read-out recorder.

This design is characterized by great stability and accuracy. Both the sample and reference beams follow a common optical and electronic course over most of the route from source to detector, and are physically separated only in the vicinity of the sample compartment. The simultaneous amplification and electronic comparison of two different frequencies of the chopped signals has some advantages over the alternative technique of synchronous rectification of a single alternation resulting from the switching of a beam between sample and reference channels.

The prism has a base 100 mm on a side; the grating is 100 × 127 mm and is ruled with 75 lines per mm.

The slits and scanning controls are programmed to permit spectral recording either at constant resolution, at constant energy, or at manually chosen slit openings.

A multipot zero compensation circuit is part of the design, so that the base line may be smoothed out as desired at 22 points over the entire range. This permits a flat base line to be obtained at all times, even when using fogged or dirty cells.

The accuracy of the instrument is ± 0.5 cm^{-1} in wavelength; $\pm 0.1\%$ in transmittance. Resolution is 0.3 cm^{-1}; stray light is 0.1% T or less over the entire range; and the drift in the base line is less than 0.1% T per day. The cabinet is thermostatted, and an internal blower-treater unit removes CO_2 and controls the

(*Continued on page A72*)

humidity of the atmosphere within the instrument.

Unicam

A well-designed instrument of the double beam, optical null balance type is the Model SP. 100 Infrared Spectrometer manufactured by Unicam Instruments, Ltd., Cambridge, England (available in the U. S. through Wilkens-Anderson Co., Chicago 51, Illinois). This instrument is available with a standard single-pass, Littrow monochromator employing a 60° prism, or with a prism-grating monochromator. The optical design of the latter type of instrument (Model SP. 100 with SP. 130 grating accessory) is shown in Figure 27. This design, in which the grating is an accessory which is attached to a standard spectrometer, has the advantage that the prism/grating combination can be employed when the highest resolution is desired, as in searching for minor constituents, or in studying the shapes of bands; and yet the grating can be removed and the instrument used as a prism spectrometer when an increased signal-to-noise ratio is needed, as in routine analyses.

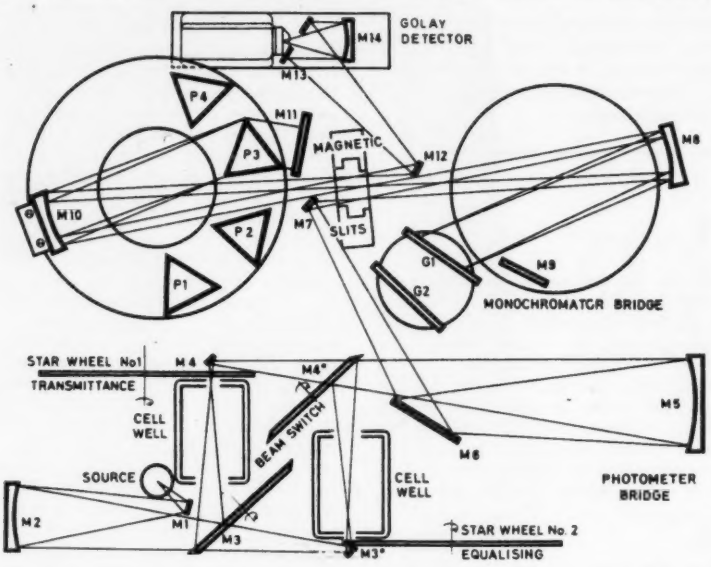

Figure 27. Optical diagram of the Unicam Model SP.100 Infrared Spectrometer with Model SP.130 Grating Accessory.

The prism is mounted on a turntable with space for up to 4 prisms of different material. Interchange of prisms and their associated cams is made by push-button, and an illuminated panel indicates the components in service.

Two of the unique features of this spectrometer are the magnetic slit control, and the starwheel attenuator. The magnetic slits consist of two plates which can be moved relative to one another. They are suspended on spring-loaded steel tapes which are fixed at their lower ends to the frame of the unit and at their upper ends to two strong leaf springs. Adjustable tension is provided by additional coil springs. One plate, on which the right-hand slit jaws are mounted, is pulled to the right by a loudspeaker type coil operating in a suitable magnetic gap at the same time as the other plate on which the left-hand slit jaws are mounted is pulled a similar distance to the left by another coil. The coils are connected in series, and are in a cathode follower circuit which follows

(*Continued on page A74*)

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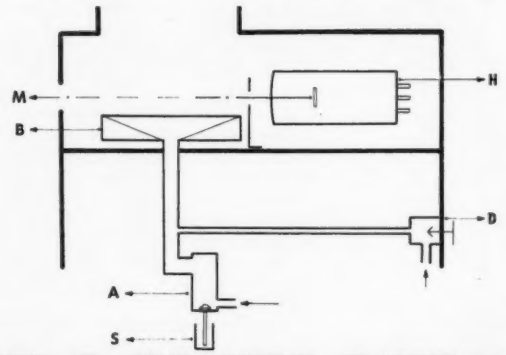

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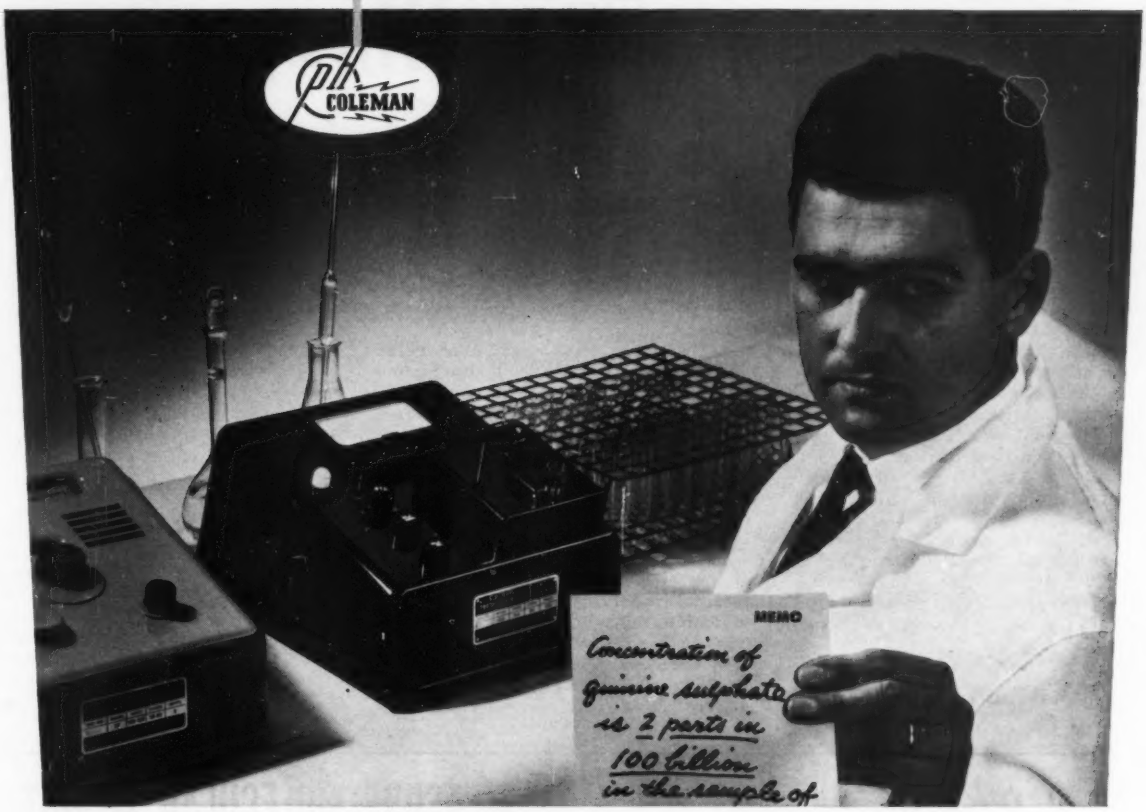

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The starwheel attenuator consists of a toothed wheel roating at 3000 rpm, which is moved into, or out of, the radiation beam in proportion to the light intensity in that beam. The starwheel has 24 spiral-cut teeth, so constructed that a radial movement of 50 mm causes the mean transmission of light through the wheel to vary linearly from 0% at the base to 90% a few millimeters from the end of the teeth. This type of attenuator is used in lieu of the comb type described previously to avoid any possible error arising from masking only part of the source image if it is not homogenous. A starwheel with an attenuator range of 5 to 15% transmission is mounted in the sample beam, and is adjusted manually by the operator. A starwheel with an attenuator range of 0 to 100% transmission is mounted in the reference beam, and is adjusted automatically during a scan by the servomechanism.

The monochromator is thermostatted, and the entire optical path is evacuated, or may be flushed. Evacuation of the optical system prevents the overloading of the servo system which tends to occur in regions of strong atmospheric absorption. A rotary oil pump and oil diffusion pump are integral parts of this spectrometer.

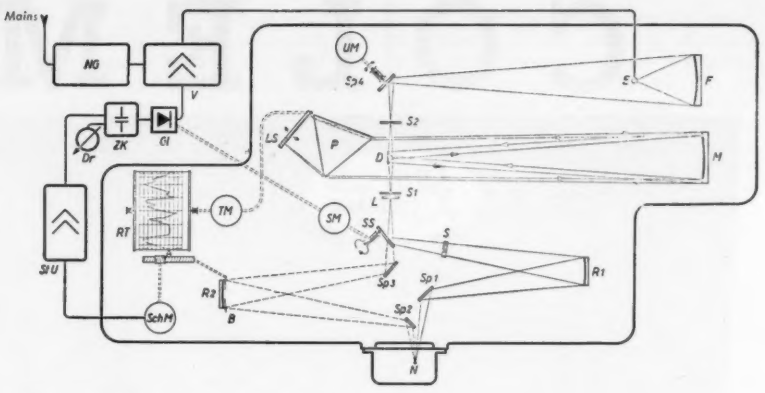


Figure 28. Optical diagram of Leitz Infrared Spectrograph.

The grating accessory consists of two gratings, with ruled areas of 90 × 60 mm, which are mounted back-to-back, slightly out of parallel. Fine controls on the grating mounting allow an independent rotational adjustment of each grating about a horizontal or vertical axis. The whole grating assembly is mounted on a turntable, and frequencies are scanned by rotation of this table through a lever and cam mechanism. One grating is ruled with 1500 lines per inch, and is used for the range 650 to 2150 cm^{-1}; the other grating has 3000 lines per inch, and covers the range 2150 to 3650 cm^{-1}.

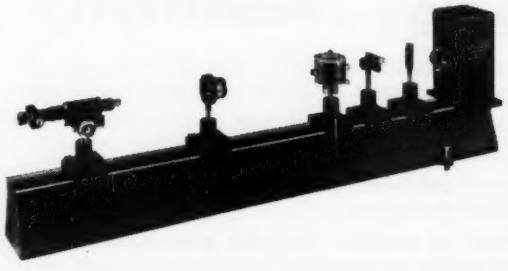
Resolution is 1 cm^{-1} at 900 cm^{-1} for the prism spectrometer, and 0.3 cm^{-1} for the prism/grating instrument. Transmission reproducibility is ±0.2% T for the prism instrument, and stray light is less than 2% at 700 cm^{-1}.

Leitz

The Leitz Infrared Spectrograph, manufactured by Ernst Leitz, Wetzlar, Germany, and available in the U. S. through E. Leitz, Inc., New York, N. Y., is a double-beam, single monochromator, optical null balance instrument of conventional design. The optical circuit, shown in Figure 28, is essentially the same as that of the Baird-Atomic Model NK-1 (*cf.*

(*Continued on page A76*)

Direct Measurement of Difference in Refractive Index



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This instrument is listed in U.S. Government specifications for the evaluation of certain clinical materials.

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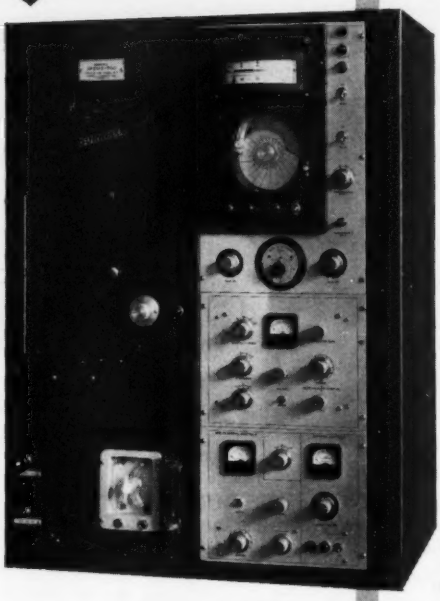

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Figure 8), or the Perkin-Elmer Model 21. The beam intensity in the reference channel is adjusted by means of the diaphragm, *B*, until the reference and sample beams are equal. The recorder pen is linked mechanically to the motion of the reference diaphragm. The source, *N*, is a Nernst glower, and the detector, *E*, is a thermocouple.

Resolution is 1 cm^{-1} at 10 microns with the 60° NaCl prism, 150 mm base, 100 mm height. Accuracy of wavelength setting is ±0.015 micron in the NaCl range, and accuracy of transmittance measurements is ±1% of the transmittance for values above 20% T.

Next: Nuclear Radiation Detectors.

Requests have been received for additional detailed description of the Coleman Flame Photometer. The following section supplements the general treatment of flame photometers which appeared in the November, 1960, issue.

10. Flame Photometers (Supplement)

Coleman

Coleman Instruments, Inc., Maywood, Illinois, manufactures a very popular instrument, designated as the Coleman Model 21 Flame Photometer. This is fundamentally a single-beam, filter-photometer instrument. The atomizer-burner unit, shown in Figures 16 and 17, is designed to produce a stable, concentrated flame, into the base of which the sample is sprayed. The fuel gas, which may be either city gas (natural or manufactured), butane, or propane, is introduced through the support tube. Its flow pattern, created by the shapes and positioning of the baffle plate and Teflon flame spreader, is such that it mixes rapidly and uniformly with the oxygen gas coming up from the atomizer tube. A combination

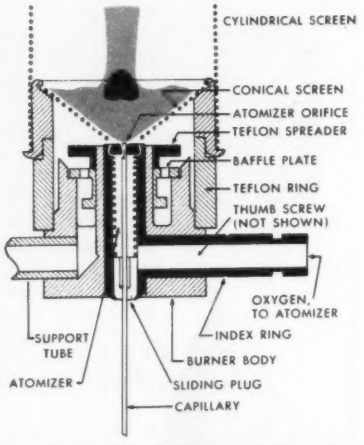
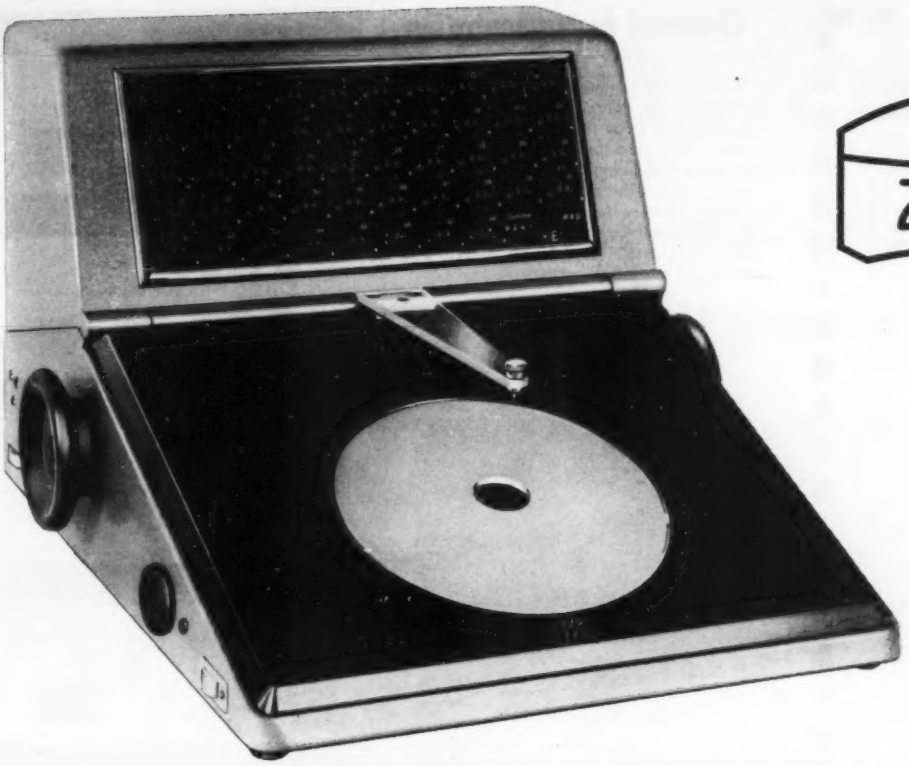


Figure 16. Diagrammatic representation of the design of the burner-atomizer unit of the Coleman Model 21 Flame Photometer.

(*Continued on page A78*)



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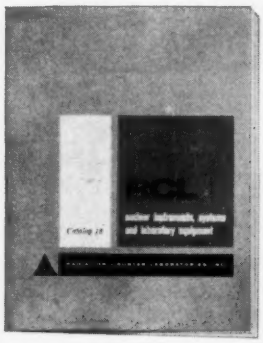

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of a conical and a cylindrical screen serves to localize and control the volume of the flame, as well as to protect the flame from drafts. The streaming of the oxygen gas through the atomizer tube creates the suction that pulls the sample up through the capillary (the bottom end of which dips into the sample container), and leads to the atomization of the sample at the top opening of the capillary.

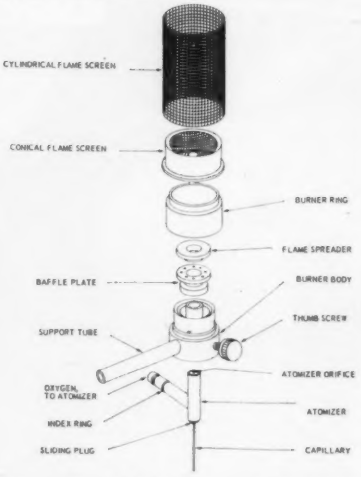


Figure 17. Exploded view of the burner-atomizer unit of the Coleman Model 21 Flame Photometer.

The light generated in the flame is directly viewed by a vacuum phototube, through a filter to isolate either the sodium, potassium, or calcium emission lines. The photometric circuit is shown in simplified schematic form in Figure 18.

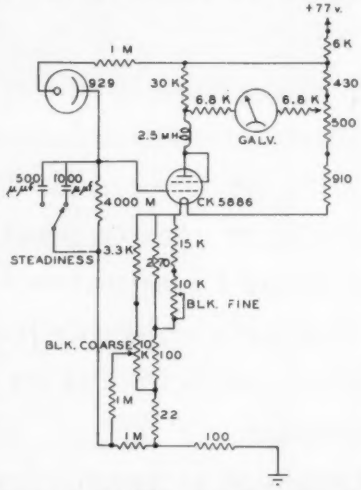
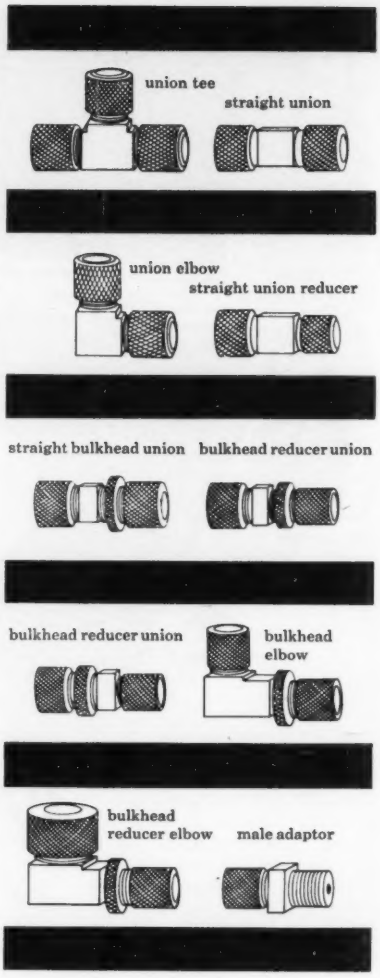


Figure 18. Simplified schematic of the photometer circuit employed in the Coleman Model 21 Flame Photometer.

The light impinging on the phototube cathode causes electron emission, resulting in a flow of electrons up through the 4000 megohm resistor. This generates an IR-drop, or voltage difference, across that

(*Continued on page A82*)



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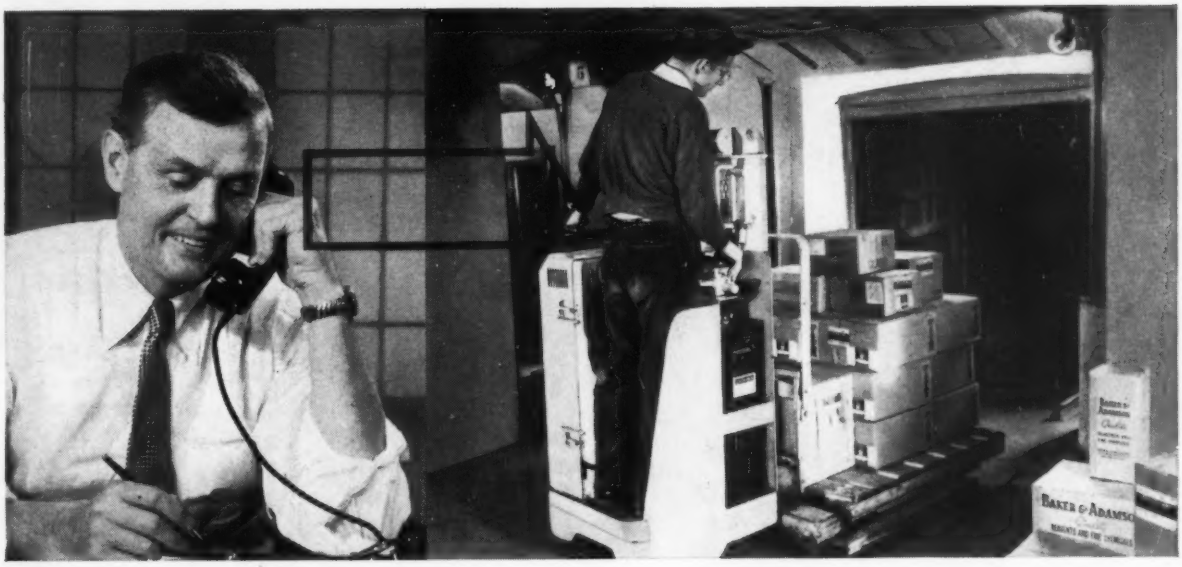

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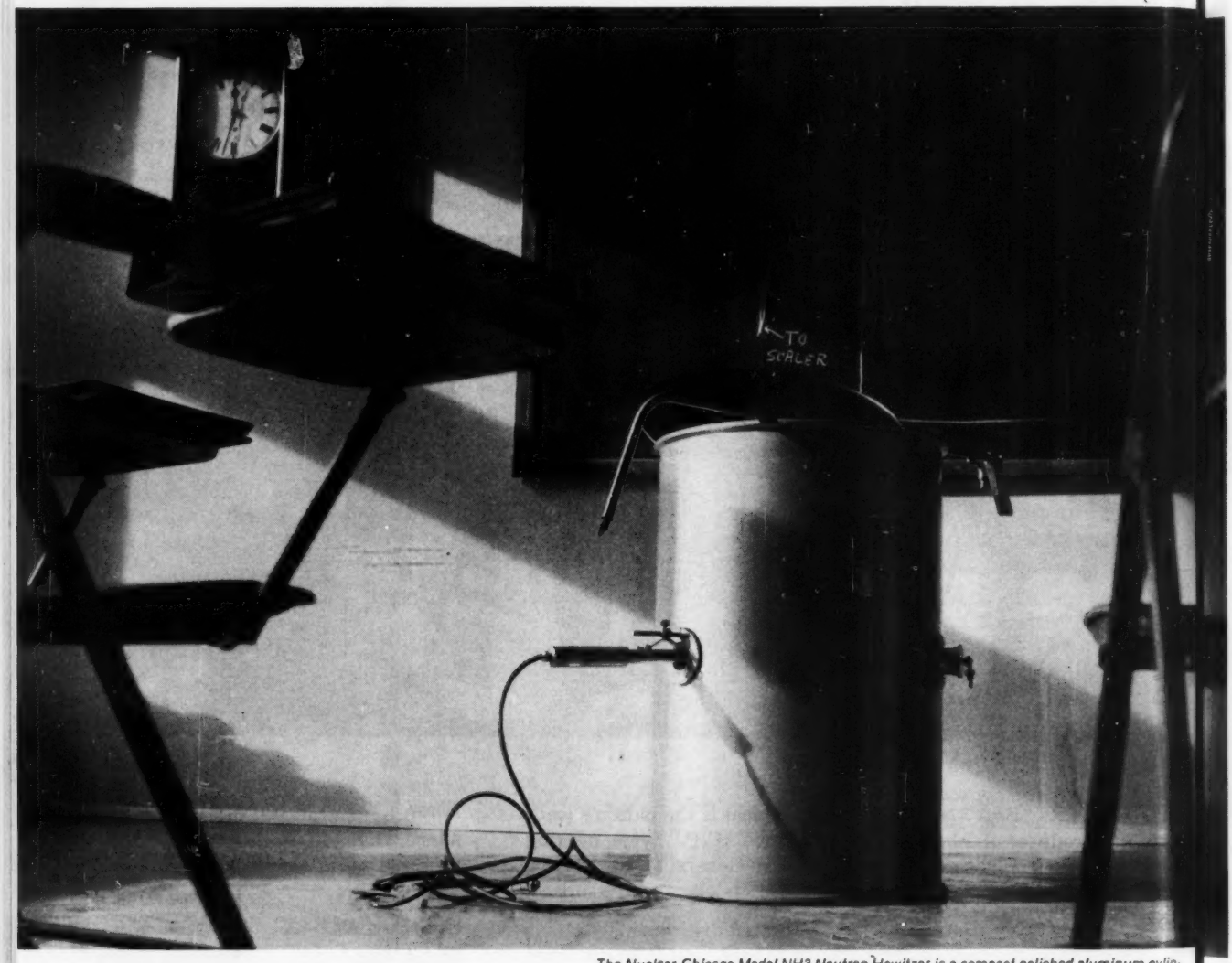
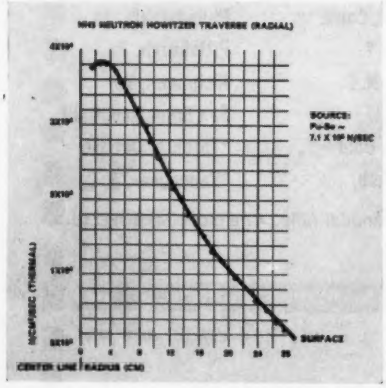


The Nuclear-Chicago Model NH3 Neutron Howitzer is a compact polished aluminum cylinder 22½" in diameter and 35" high. Ports are provided for neutron activation experiments.



Thermal neutron flux distribution in entry port using 5 curie plutonium-beryllium source. Maximum flux is nearly 4 x 10^4 n/cm^2/sec about 3 cm from the center line. Reasonably large samples can be irradiated with only slight flux variation within 6 cm radius from the source.

Physical Description of the Howitzer. A central storage tube in the Neutron Howitzer accepts as many as five one-curie plutonium-beryllium sources. Neutrons emerge from the source with average energies in the 4 MEV range. A moderating mass of paraffin around the central tube slows these energetic neutrons down to "thermal" velocities (approximately 0.02 EV). Two horizontal entry ports extend from the central tube through the paraffin to the outside of the vessel. The chart at the left shows the radial thermal flux distribution.

The neutron flux can be measured with BF_3 detectors, scintillation detectors, or with foil activation methods which are described in the experiment manual. Detector mounts, port drawers, indium and cadmium foils and a variety of foil holders and spacers are provided to simplify such studies.

Radioisotope Production and Activation Analysis. Radioisotope production and activation analysis are dramatic uses of the Neutron Howitzer which can stimulate and intrigue the student. Although the thermal neutron flux in the howitzer is relatively low when compared to reactors or particle accelerators, there are many elements which can be activated in a reasonable time in the device. Radioactive isotopes produced in the howitzer are generally of low intensity, requiring only normal care in the handling of the activated samples.

Activation analysis with low-level sources has one major advantage over activities produced in a high flux device such as a reactor or accelerator. Only isotopes with a high activation cross section and a short half-life can be detected after a short irradiation, minimizing the confusion of multiple neutron-induced activities.

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Now, for the first time, a commercial instrument is available for detailed study and experimentation with neutron sources. Although substantial progress has been made in the education of students in the atomic energy field, the demonstration and study of neutron radiations and effects has been largely neglected. To fill this important educational need, Nuclear-Chicago Corporation conceived and designed the Model NH3 Neutron Howitzer.

The NH3 Neutron Howitzer is a compact, versatile device designed for effective laboratory and lecture use in colleges and universities. A detailed manual supplied with the howitzer contains explicit and carefully tested experiments which carry the inexperienced student safely and naturally from simple radiation measurements to relatively complex studies of neutron reactions. The design of the howitzer is such that original experiments and research can be easily carried out.

Supplied with all necessary accessories, the NH3 Neutron Howitzer from Nuclear-Chicago forms a particularly appropriate nucleus for new radioisotope laboratories and makes an outstanding low cost addition to existing educational facilities. For complete information please write for our NH3 Brochure.

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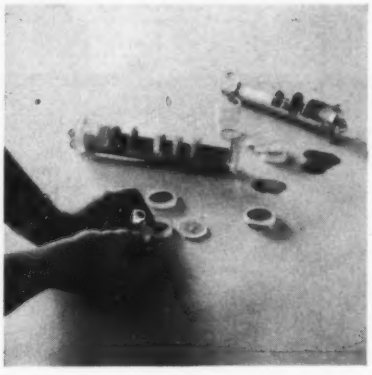
The Neutron Howitzer Laboratory. The average laboratory using radioactivity already has most of the instruments required for optimum use of the Neutron Howitzer. A simple preamplifier converts any Nuclear-Chicago scaler for use with BF_3 neutron detectors. Foils and samples emitting gamma rays can be counted with highest efficiency in a well-type scintillation detector and a combination scaler-pulse height analyzer. If continuous recording of gamma-ray spectra is desired, a recording spectrometer is invaluable.

Most irradiated samples also emit beta radiation and can be measured with simple beta counting instrumentation. An end-window Geiger tube, detector mount, scaler, and timer are required. An absorber set will permit calculation of beta energies and will also help in isotope identification.

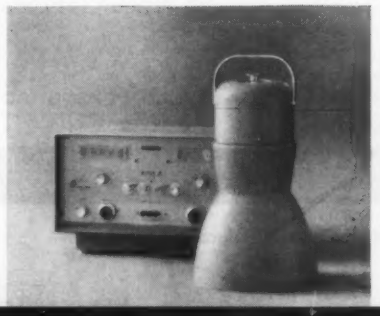
42 inch source handling tong is provided for manipulation of neutron sources.



A foil is placed in a holder for activation. A variety of foils are supplied with the howitzer.

Spacers are provided for the port drawers so the holder can be positioned at various radii from the source.

A detector mount is furnished for each port. BF_3 detectors can be mounted perpendicular to the port opening or inside the port.



Nuclear-Chicago Model DS5-5 Scintillation Well Counter and Model 132A Analyzer Computer provide a means of identifying gamma emitting samples by energy discrimination.

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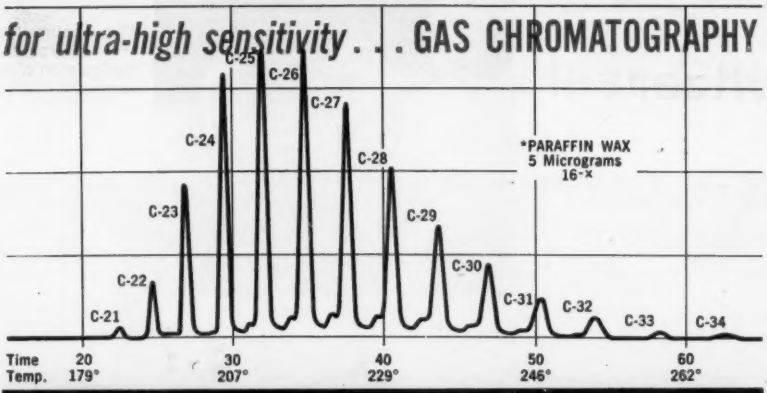

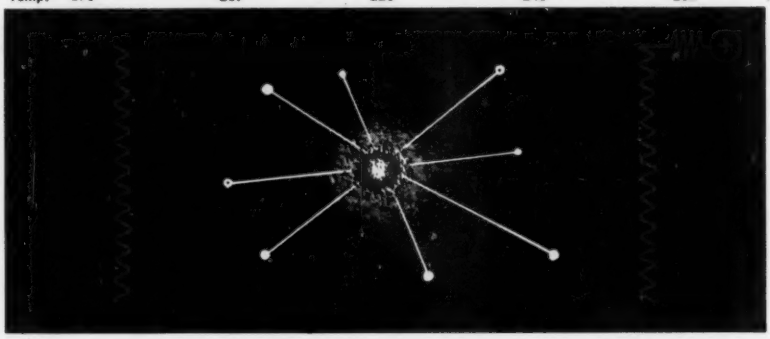

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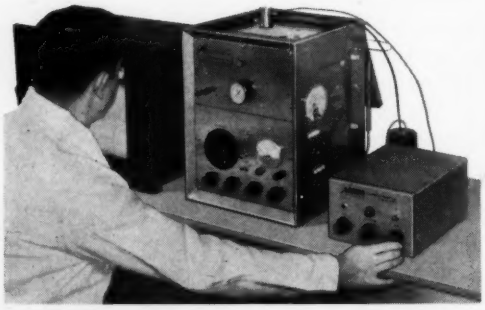

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resistor, and this signal serves to control the grid-to-cathode voltage difference in the CK5886 electrometer tube. Consequently, the current reaching the plate of this tube is a measure of the light intensity seen by the phototube.

The capacitor in parallel with the 4000 megohm phototube resistor introduces a time delay in the building-up, or decay-away, of the IR-drop across that resistor due to the flow of photocurrents. Hence, it serves to smooth out the response of the electrometer tube when the light output of the flame is subject to rapid fluctuations in intensity; i.e., it results in an effective averaging of the flame intensity over a time period that is determined by the time constant (= R × C) of the phototube circuit. The two capacitors provided on the *"steadiness"* selector switch permit the time constant to be changed by a factor of 2 to adjust to different degrees of flame stability. The *"blk"* controls serve to adjust the zero setting of the galvanometer to some convenient scale position, usually zero, when there is no sample, or a blank, in the flame.

The output of the photometer may be read on any low resistance meter having a sensitivity of 1 microampere or less for full-scale deflection. The galvanometer is not provided with this instrument; instead, a plug-in connection permits this flame photometer to utilize the galvanometer read-out of any of the Coleman colorimeters or spectrophotometers, or to be connected to any other suitable galvanometer.

Samples are atomized into the flame at the rate of about 1 ml per minute, and a stable reading is obtained in 10–20 seconds. Full-scale deflections are obtained with solutions containing 1.2 ppm of sodium; this corresponds to a sensitivity of detection of sodium of about 0.02 ppm, under conditions when a signal 2% of full-scale can be detected over the flame background.

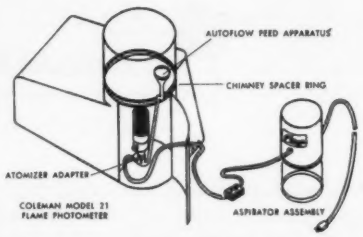

Figure 19. Design of the Coleman Autoflow Sample Handling System for getting maximum utilization of small volumes of sample.

A useful accessory available with this instrument is the Autoflow Sample Handling System ($76), the principle of which is shown in Figure 19. The glassware and aspirator pump make possible maximum utilization of a small volume of sample; as little as 0.2 ml of sample will give 12 seconds of galvanometer deflection with this method of introduction of the sample into the flame.

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EASY TO USE *...anyone can weigh quickly and accurately with the Right-A-Weigh ... frees skilled people for other work.*

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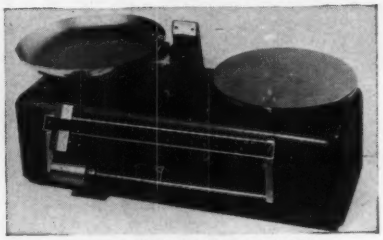

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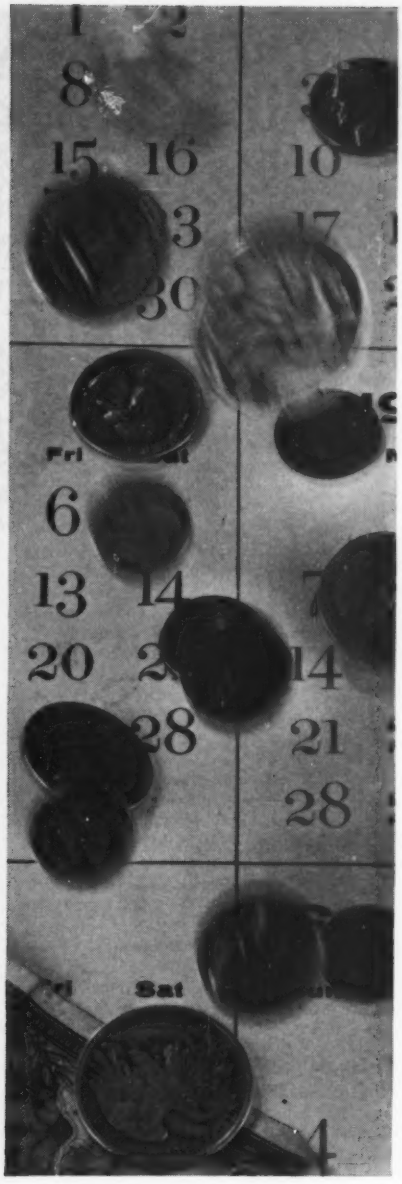

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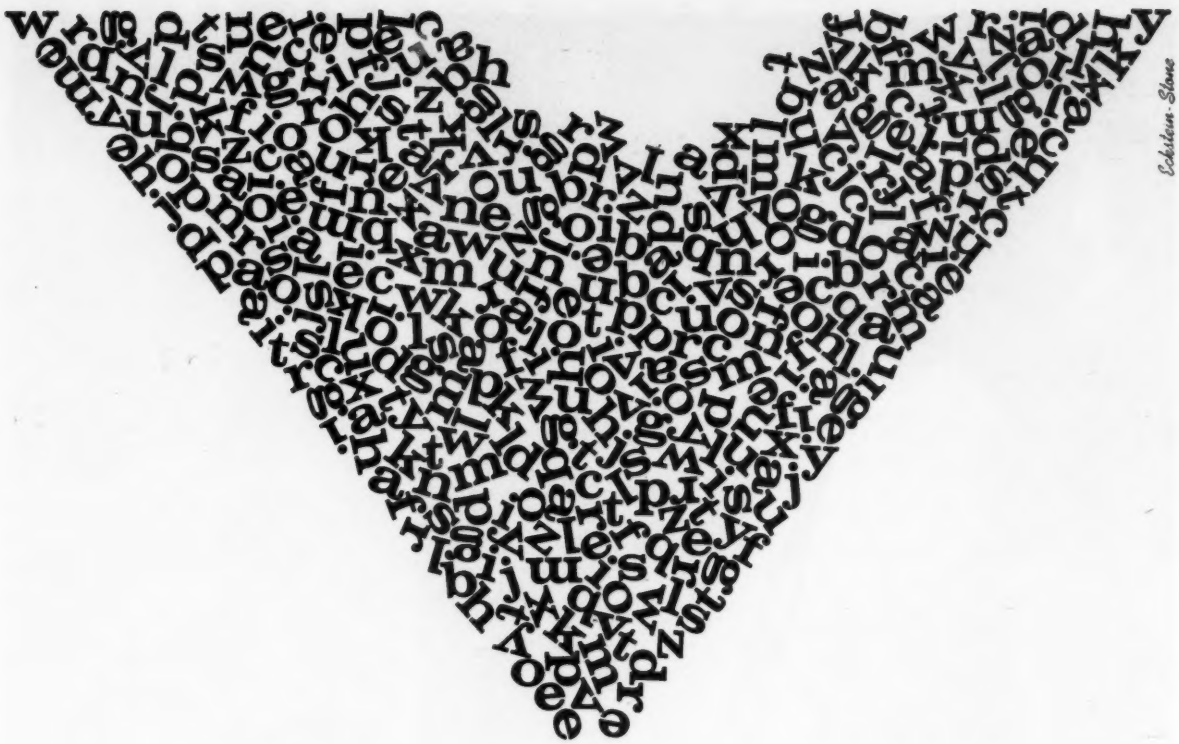

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The analyst who has never been besieged with laboratory filtration problems just doesn't exist. Nor does a filter paper exist with the magical property of being all things to all analysts. Even Reeve Angel does not yet have the panacea of filter papers. But, we do offer a vast selection in a diversified line to meet almost every need you are likely to encounter—and a skilled technical staff who can make intelligent, knowledgeable recommendation on your specific filtration problems. We invite your inquiries. Write today for a list of Reeve Angel filter products.

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Chem Ed

Out of the EDITOR'S BASKET

When writing for additional information about new products or for new literature, your inquiry will receive prompt attention if you write on your firm or institution letterhead and mention the Journal of Chemical Education and the date of the particular issue. Pamphlets, booklets and similar literature are gratis unless otherwise specified.

New Apparatus and Equipment

▶A new miniature $^1/_{12}$ HP oil-less motor-powered compressor and vacuum pump that is said to deliver more free air than any rotary, diaphragm or piston design unit of the same horsepower is now available from Bell & Gossett Co., Morton Grove, Illinois.

▶The new Model 2586-S "Cutie Pie," recently announced by Nuclear-Chicago Corporation, 359 E. Howard at Nuclear Dr., Des Plaines, Illinois, is the first gun-type survey meter which provides both rate of dose and accumulated dose measurements. The instrument is used for monitoring beta, gamma, and x-radiation of low and medium intensity.

▶ Conventional Buechner Funnels, which are made in one piece, make cleaning a severe problem. The American Agile Corp., 5461 Dunham Rd., Maple Heights, Ohio, offers a new Buechner Table Type Funnel which features a removable filter plate section for ease of cleaning.

▶ Lourdes Instrument Corp., 656-76 Montauk Ave., Brooklyn 8, N. Y., has developed a unique metal support and sealing cap (patent pending). It permits fragile polyethylene bottles to be completely filled and effectively sealed for centrifugation in angle type, superspeed centrifuges at forces up to 68,000 × Gravity.

▶ The Dec-O-Gram, a new high-form heavy duty triple beam balance, has been introduced by Ohaus Scale Corp., 1050 Commerce Ave., Union, New Jersey. High capacity (2610 grams), fine sensitivity, and sturdy construction, highlight this newest addition to the Ohaus line.

▶ Yellow Springs Instrument Co., Box 106, Yellow Springs, Ohio, announces the release of a new price dual-channel recorder, the YSI Model 81. The Model 81 provides two channels of input which simultaneously record full scale. Trace identification is provided to avoid confusion when recorded parameters cross or run close to each other. The unit can be run either dual-channel or single-channel as required.

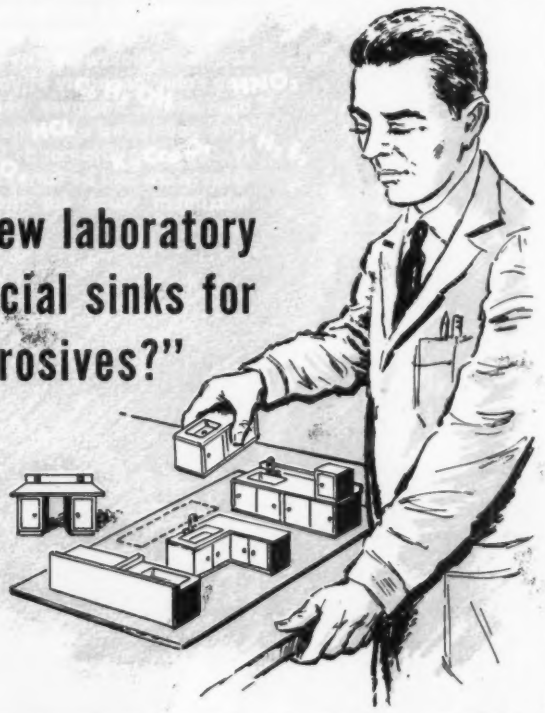
▶ A Stainless-steel vacuum trap, Vactrap, designed to provide a handy portable condensing system with ultimate vacuum of .3 microns, has been introduced by Instrumentation Associates, 17 W. 60th St., New York, N. Y.

▶ A Threaded-Glass and Teflon Needle Valve which provides fine fluid control and permits continuous visual observation of flow has been introduced by the Glass Products Division, Fischer & Porter Co., Warminster, Pennsylvania. The glass tube itself is threaded by an exclusive F&P process which eliminates the need for conventional metal threads and allows for a simplified two-piece construction.

▶ To meet the many and varied demands of laboratory and industrial users, Hoke Inc., 1 Tenakill Park, Cresskill, New Jersey, has developed an entirely new line of general purpose pressure regulators. Designated as the 680 Series, these regulators provide a control accuracy of 2%. Delivery pressures to 140 psi, and rated flows to 2000 SCFH air are obtainable with excellent regulation characteristics.

▶ Butyl dry box gloves are now available that are said to be as much as 10 times less permeable to gases and moisture as

(*Continued on page A89*).



"Will our new laboratory require special sinks for special corrosives?"

Not if you are careful to specify *U. S. Chemical Porcelain Sinks*. For these are the laboratory sinks with time-proven ability to handle all corrosives safely.

Years of continuous service in leading hospitals, universities and industrial laboratories throughout the world attest to the everlasting durability of solid chemical porcelain under all types of corrosive conditions. Acids, alkalies, caustics, solvents —hot or cold, mixed or concentrated—a "U. S." sink handles them all. Commonly used cleaning solutions (such as potassium chromate and concentrated sulphuric acid) which may attack other materials cannot harm "U. S." porcelain sinks. With normal routine rinsing even hydrofluoric acid presents no special problem.

"U. S." sinks offer many excellent advantages. Scratch-resistant, stain-free, smooth gray-glazed surfaces assure a lifetime of beauty. No honing or scouring is ever required. High mechanical strength and superior heat-shock resistance for years of reliable, maintenance-free service.

Your Laboratory Furniture Dealer can give you complete details. See him or write for your free copy of new Bulletin L-8R describing over 50 standard sizes and many styles.

CHEMICAL CERAMICS DIVISION

U. S. STONEWARE

AKRON 9, OHIO

121-G

OOPS!

Yes . . . accidents can and will happen as long as glassware is used by students or lab technicians. And it always seems to happen to the most expensive item!

If you have been buying expensive heat-resistant laboratory glassware under the impression that it is stronger, more rugged and less prone to break . . . stop and think for a minute. Does this really make sense?

If you intend to use the item in contact with a flame, then purchase heat-resistant glassware, by all means. If you value economy in your laboratory operations, however, think twice before you spend extra money needlessly. Specify Diamond D Laboratory Glassware and be sure you are getting maximum value for the money you spend.

DOERR GLASS COMPANY
Vineland, New Jersey

Out of the EDITOR'S BASKET

existing neoprene gloves. For additional information, write CAEMCO P. O. Box 3693, Jacksonville 6, Florida.

▶ Scientific Glass Apparatus Co., Inc., Bloomfield, New Jersey, announces the development of a new device called Burd-Watcher II for automating, programming, monitoring, and controlling reactions and distillations of *any* size. According to the company, the versatility of the Burd-Watcher allows almost endless variations in circuitry to fit individual programs needed for chemical reactions and distillations.

▶ U. S. Nuclear Corp., P. O. Box 208, 801 N. Lake St., Burbank, California, designs and builds irradiators employing Co-60 and Cs-137 sources. Cavity size and materials have been selected to allow irradiation of diverse systems ranging from small animals and other biological samples to dynamics of chemical and electronic experiments to testing of materials. Design is such that continuous instrumentation of experiment and control of environment is possible.

▶ Alconox, Inc., makers of laboratory detergents have announced a new 50-Dispenser Pack. The dispenser, with convenient tab for hanging on the wall, contains 50 individually-sealed $^1/_2$ oz. packets of Alconox, each measured to make $^1/_2$-gallon of the detergent that meets the most exacting requirements of hospital, medical and laboratory use and is recommended for the safe, thorough cleaning of all instruments, glassware and equipment. The new dispenser pack may be ordered from any laboratory supplier.

▶ The Professional Tape Co. is now making a new scientific tape called "TSI Tape," the positive reactor tape constructed to react at 250°F. after a 15 minute period in the autoclave or oven. It is a pure white tape and the word "Sterile" shows after sterilizing. Because of its scientific calibration, it is most adapted for sterilizing glassware and can be attached to any solid material such as glass beakers, trays, Petri dishes, wire baskets, cannisters, etc.

TSI (Time Sterile Indicator) Tape is available in a continuous roll either in 500″ or 2160″ lengths. Complete details and prices are in a folder available from Professional Tape Co., Inc., 355 Burlington Rd., Riverside, Illinois.

New Literature

● A new melting point apparatus which for the first time provides reproducible results of known and constant accuracy is now available. For the fully illustrated four-page bulletin (*No. 322*) write to Harshaw Scientific, Division of The Harshaw Chemical Co., 1945 E. 97th St., Cleveland 6, Ohio.

(*Continued on page A90*)

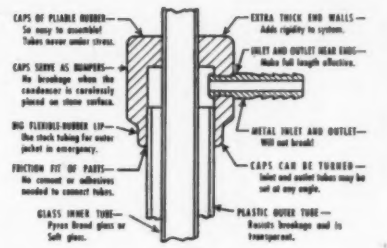
WELCH "Straight Jacket" CONDENSERS

Now Feature Outer Jacket of Unbreakable Transparent Plastic

Metal inlet and outlet tubes can be placed at any convenient angle

Actual use will convince you of the superior features of the Welch Straight Jacket Condenser

No. 4941.


CAPS OF PLIABLE RUBBER— So easy to assemble! Tubes never under stress.
CAPS SERVE AS BUMPERS— No breakage when the condenser is carelessly placed on stone surface.
BIG FLEXIBLE-RUBBER LIP— Use stock tubing for outer jacket in emergency.
FRICTION FIT OF PARTS— No cement or adhesives needed to connect tubes.
GLASS INNER TUBE— Pyrex brand glass or Soft glass.
EXTRA THICK END WALLS— Adds rigidity to system.
INLET AND OUTLET HEAD ENDS— Make full length effective.
METAL INLET AND OUTLET— Will not break!
CAPS CAN BE TURNED— Inlet and outlet tubes may be set at any angle.
PLASTIC OUTER TUBE— Resists breakage and is transparent.

PLASTIC OUTER JACKET—Soft glass tube

Complete with soft glass inner tube and plastic outer tube.

Length of jacket, mm,	250	300	400	500	600	750
Length over-all mm,	475	525	625	725	825	975
O.D., outer tube, mm,	25	25	25	25	25	25
O.D., inner tube, mm.	12	12	12	12	12	12
No. 4941 Each,	$1.45	$1.55	$1.65	$1.85	$2.00	$2.70

PLASTIC OUTER JACKET—Pyrex-brand glass tube

Complete with Pyrex-brand glass inner tube and plastic outer tube.

Length of jacket, mm,	300	400	500
Length over-all, mm,	525	625	725
O.D., outer tube, mm,	25	25	25
O.D., inner tube, mm,	12	12	12
No. 4941P. Each	$2.55	$2.80	$3.10

THE WELCH SCIENTIFIC COMPANY

ESTABLISHED 1880

1515 Sedgwick Street, Dept. D-1, Chicago 10, Illinois, U. S. A.

Manufacturers of Scientific Instruments and Laboratory Apparatus

SUPER-COLD

with EXTRA-FAST PULL-DOWN TIME!

Forma

FRIGID-MIDGET

The Frigid-Midget has a cold walled, stainless steel inner work chamber, (fast pull-down time from ambient to −100°F in 1¼ hrs). Air-circulation fan housed in separate compartment with safety perforated grille. Separate switch to cut off air circulation when lid is open.

Inner chamber entirely insulated..Special glass wool around heat exchanger..Double-gasketed with silicone rubber..All controls front mounted including access panel for through connections.

TEMPERATURE RANGE −100° F to ambient. (Other ranges avail.)

OVERALL DIMENSIONS—24″ W × 23″ F. to B. × 38″ H.

INSIDE DIMENSIONS—12″ W × 14″ F. to B. × 12¾″ H.

115 Volts *11 Amps* *60 Cycles*

ECONOMICAL
Capable of handling work normally done by far more costly equipment!

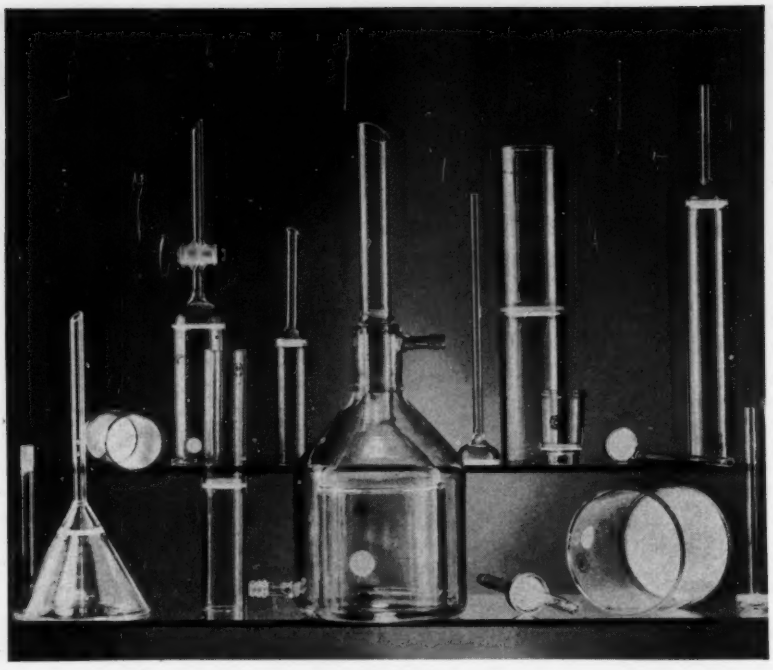
VERSATILE
So many uses, no laboratory can afford to be without a Frigid-Midget! These include testing of electronic components, instruments, metal-aging, shrink fitting, tool fitting.

MOBILE
Special added features are its compactness and mobility.

Cat. No. JC 70493..Each, $1600.00

STANDARD SCIENTIFIC Supply Corp. 808 BROADWAY NEW YORK 3, N.Y.

LABORATORY APPARATUS REAGENTS AND CHEMICALS



WHY SELECT ACE FIBER GLASS SINTERED FILTERS?

Quality and Greater Abrasion Resistance. Ace filters, the first American made sintered glass filters, feature a glass fiber structure, more abrasion resistant because it is fused together on a larger area. Particles do not detach from the filter body as easily as spherical granules. The shock and chemical attack resistance of glass is unimpaired as the Ace fiber glass sintered filter is made entirely of glass. You are assured of Ace Glass quality: each filter plate is *individually* tested for porosity and hardness.

Selection and Economy. Ace fiber glass sintered filters are economically priced. For instance, the Filter Funnel (Cat. #7305 in the 20 ml. cap. with 20 mm. disc), shown in photograph above, is listed at $3.20. Ace fiber glass sintered filters have been incorporated into a wide variety of Ace glassware described in our new filterware brochure. Get it for your files!

Write Dept. J for your copy of Filterware Brochure No. 6050.

ACE GLASS INCORPORATED

Louisville, Ky., Vineland N. J. Springfield, Mass.

Out of the EDITOR'S BASKET

● Write to Schuco Scientific, 75 Cliff St., New York 38, N. Y., for complete catalogue of "Heto" pumps. They are adaptable to a variety of different tasks.

● *Data Sheet 51*, describing a new one-cubic foot capacity Glasteel conical dryer-blender, is available from The Pfaudler Co., a division of Pfaudler Permutit Inc., 1122 West Ave, Rochester 3, New York.

● Just off press and packed with hundreds of unusual and hard-to-get values in Science, Math and Optical equipment, a new 144-page Catalogue is now available from Edmund Scientific Co., Barrington, New Jersey.

● Complete technical and operating data on Cenco's Color Matic Endpoint Detector and Digital Readout Volumatic Syringe has been published by Central Scientific in a new 16-page booklet. *Booklet 285* is available from Central Scientific, a division of Cenco Instruments Corp., 1700 Irving Park Rd., Chicago 13, Illinois.

● A new 40-page booklet giving complete chemical and physical properties of caustic soda is available from U. S. Industrial Chemicals Co., 99 Park Ave., New York 16, N. Y.

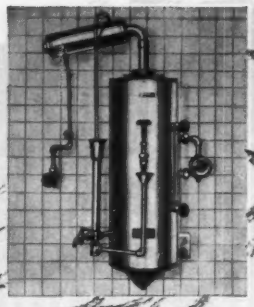
● Atomic Energy of Canada Ltd., P. O. Box 93, Ottawa, Canada, offers a fine new *Catalogue D* describing industrial and medical products and services available. A companion volume which includes a variety of general and technical information is entitled, *The AECL Radioisotope Handbook*. Request them on your letterhead.

● A new 24-page *Catalogue J1060* provides data on the complete line of unbreakable, corrosion-resistant, plastic laboratory ware manufactured by the Nalge Co., Inc., Rochester 2, N. Y.

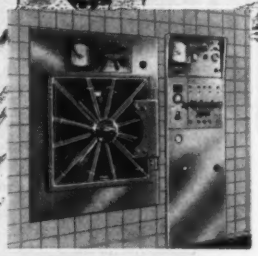
● Mann Research Laboratories, Inc., 136 Liberty St., New York 6, N. Y., offers their reference guide and price list *No. 126* of *Mann Assayed Biochemicals for Research*.

● Antara Chemicals, 435 Hudson St., New York 14, N. Y., has published a ready-reference data sheet *#AP-60-3* on all of the company's commercially available high-pressure acetylene derivatives. Each of these products, which include PVP-VA Copolymers, PVP-Iodine, Vinylpyrrolidone, Methylpyrrolidone, 2-Pyrrolidone, Butyrolactone, 1,4-Butanediol, Butenediol, Butynediol, and Propargyl Alcohol and Halides, is described in considerable detail.

● A new catalogue of chromatography equipment *CP-60*, has just been released

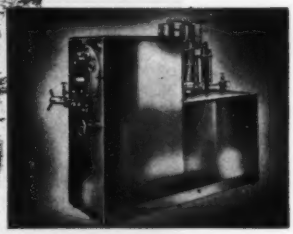


WATER STILLS
Capacities from ½ to 500 gal./hr.
Steam, Electric or Gas Heat.



STERILIZERS
Pressure Steam, Gas-Steam,
and Gas. Chambers 16″ x 16″ x 24″
to 60″ x 66″ x 120″.

TALK MICROBIOLOGICAL EQUIPMENT...

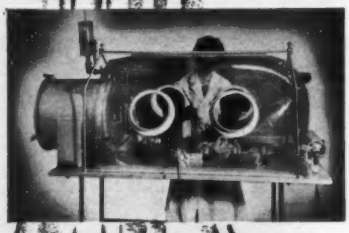


BIOGEN • *continuous culture apparatus*
For batch or continuous culture of pure
microorganisms in controlled conditions.

and you have to talk to American Sterilizer!

● Here at Amsco, sterilization is still of prime concern. However, to parallel the great strides of modern biological technology, we've developed a number of highly specialized microbiological devices—all designed to do their job better with less personnel time and attention and at a cost well within practical limits.

If you've a problem in this area, a letter to our Scientific and Industrial Department may lead to its economical resolution.

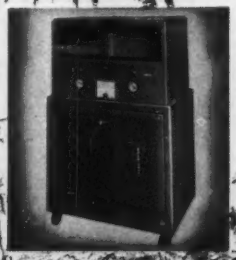


DRY BOX
Flexible film chamber.
Efficient, economical,
easy to work with.

SCIENTIFIC AND INDUSTRIAL DEPARTMENT

AMERICAN STERILIZER
ERIE • PENNSYLVANIA

World's largest designer and manufacturer of Sterilizers, Operating Tables, Lights and related biological equipment



FREEZE DRY APPARATUS
Laboratory, pilot plant and
production models. Used for the
preservation and concentration
of labile substances.

GERM FREE LIFE APPARATUS
Complete service, including
flexible operating,
rearing and transfer chamber.

Water de-mineral-ized to HIGHER Ionic Purity with

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Ion Exchange

WATER DE-MINERAL-IZERS

Ideal for School Laboratory use and Classroom demonstration.

Portable • Low Cost • A Size For Every Need

DEEMINIZER

In use throughout the world as laboratory standard. Capacity up to 5 gals. per hour. Comes complete with direct-reading electric quality-check meter. Reservoir supplies processed water for instant use. Gravity flow insures consistent quality. DEEMINIZER® CL-5-115V/60C-$39.50. Carton 6 DEEMINITE Resin Filters-$11.70.

Deem-a-Jet

The world's lowest cost, high capacity de-mineral-izer. Faucet connected—water pressure operated. Multi-jet feed. De-ion-izes tap water—fast—up to 2 gals. per min.—120 G.P.H. Use it for immediate volume needs—or intermittently. DEEMAJET® Model 128 complete with resins - $39.50. Extra DEEMINITE® Resin Refill Units-$21.00.

DEEMINAC

Fill quart bottle with tap water and invert on holding stand as shown. Operates unattended. Gravity flow—2 to 3 quarts per hour—insures consistent quality. DEEMINAC® Model SD-326 complete with metal stand, graduate and cover—$6.95. Extra Filters (A-6) $4.00. Demineralizer only, without stand, etc. DEEMINAC Model 32-6-$5.75. Other DEEMINAC models from $1.75.

AVAILABLE THROUGH LABORATORY SUPPLY DEALERS THROUGHOUT THE WORLD, OR WRITE DIRECT.

- Detailed bulletins upon request -

CRYSTAL RESEARCH LABORATORIES, INC.

29 Allyn St. Hartford 3, Conn.

(JAckson 7-3215)

Deeminizer—Deeminac Pat.—Deemajet Pat. app. for

Out of the EDITOR'S BASKET

by Will Corp., Box 1050, Rochester 3, New York.

● A new 6-page brochure, available from Nuclear-Chicago Corp., 359 E. Howard at Nuclear Dr., Des Plaines, Illinois, describes how to use nuclear gauges for rapid field determination of moisture and density in soils, aggregates, concrete, asphalt, etc.

● A six page folder describing and illustrating the complete line of light, medium and heavy duty stirrers is available from Talboys Instrument Corp., Emerson, New Jersey.

● The Institute of The Aeronautical Sciences, 2 E. 64th St., New York 21, N. Y., offers new edition of their publication *Your Career as an Aero/space Engineer.*

● A recent book by the renowned Soviet crystallographer, Prof. A. I. Kitaigorodskii, head of the Institute of Elemental Organic Compounds of the U.S.S.R. Academy of Sciences in Moscow, is now available in English translation from Consultants Bureau, 227 W. 17th St., New York 11, N. Y. It is entitled *The Theory of Crystal Structure Analysis,* $12.50.

New Chemicals and Compounds

◆ Applied Science Laboratories, Inc., 140 N. Barnard St., State College, Pennsylvania, offers Gas-Chrom, the new solid support (flux-calcined diatomaceous earth) for chromatograph columns, now available in 25 gram sizes. Kits made up of various mesh sizes and of regular (S), acid washed (A), and acid and alcoholic-base washed (P) grades are offered. With these kits the analyst can determine the type of Gas-Chrom best suited for a particular problem.

◆ Metal Hydrides, Inc., 12–24 Congress St., Beverly, Massachusetts, has available lithium aluminum tri-t-butoxyhydride, a new complex that will reduce acid chlorides to aldehyde.

◆ Eastern Chemical Corp., 34–A Spring St., Newark 4, New Jersey, offers experimentally a series of new hydroquinone derivatives such as the nitrodibutyl, nitrodiethyl and nitrodimethyl ethers, as well as the di-n-propyl ether of chlorohydroquinone.

◆ A pre-mixed liquid scintillator concentrate called Liquiflour, which eliminates weighing and stirring in the laboratory has been introduced by Pilot Chemicals, Inc., 36 Pleasant St., Watertown 72, Massachusetts.

Miscellany

★ The advisory committee to the Surgeon General of the Public Health Service

(*Continued on page A94*)

Free Chart shows safe methods of Handling Compressed Gases

This new chart, prepared by Matheson's Technical Department, shows safe methods of handling every gas available in cylinders for laboratory or plant use. Step by step illustrations show correct procedures for Receiving Cylinders, Moving, Handling Empty Cylinders, Using Gases. The 14" x 21" chart is designed for wall mounting wherever gases are received, stored or used. Write for your copy.

New Gas Catalog

Our new catalog gives prices and data on 85 compressed gases, gas mixtures and gas regulating equipment. Available on request.

The Matheson Company, Inc.

Compressed Gases and Regulators — East Rutherford, N. J.; Joliet, Ill.; Newark, Calif.

NOW! FASTER, EASIER PIPETTING

Leaves One Hand Free!

accropet

PIPETTOR provides most accurate filling and discharge!

Avoid the dangers and possibilities of contamination that are constantly present with conventional pipetting methods. You operate compact, ingenious Accropet simply and easily with one hand... enjoy accurate controlled discharge and filling at high speeds!

Practical, durable Accropet is manufactured by the Manostat Corporation of high temperature polypropylene... to withstand temperatures up to 320°F. "O" ring construction assures most accurate, positive, leak-proof action. Maximum flexibility, provided by rubber tubing connection, helps eliminate pipet breakage.

OPERATING INSTRUCTIONS: Attach outlet to pipet directly, with a short length rubber tubing, or a one-hole rubber stopper.

Accropet is then most conveniently operated by the thumb and first finger, while resting the body at the base of the other three fingers, and using these to hold it in place.

Using thumb and first finger, rotate knob up ¾ turn from its deepest position. Place tip of pipet in liquid, and draw up liquid by rotating knob upward.

To accurately control discharge of liquid, rotate knob down into body of Accropet, using thumb and first finger.

T20386 Accropet, ultramicro size. For all pipets up to and including 2/10 cc., and especially recommended for lambda-pettes. Packaged singly or in cartons of six.
Each $3.60. Pkg. of 6 $16.80

T20386D Accropet, micro size. For all pipets up to and including 2 cc, and red and white blood pipets.
Each $4.95. Pkg. of 6 $23.70

The EMIL GREINER *Co.*

20-26 N. MOORE ST., DEPT. 422, NEW YORK 13, N. Y.

Out of the EDITOR'S BASKET

recommended the following goals for the 1960–70 decade:

Determine the effects of air pollution on human health.

Determine the effects on the Nation's agricultural economy resulting from air pollution damage to animals and crops.

Find better ways of measuring the economic loss from air pollution damage to materials, and soiling, and reduced visibility.

Find better ways of measuring and identifying air pollutants at their source and in community air.

Develop better techniques for assessing meteorological factors affecting air pollution.

Learn, through research, more about the formation of new pollutants from reactions in the air.

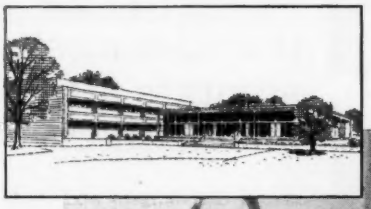
Expand our nationwide air pollution monitoring efforts.

Develop new methods and equipment for controlling the sources of air pollutants.

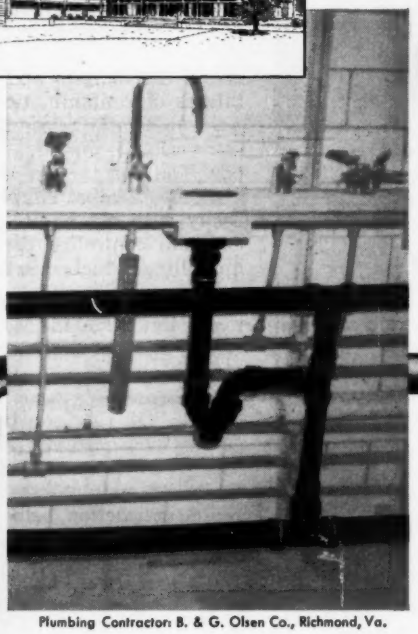
Build and disseminate a comprehensive body of knowledge related to the technical, legal, economic, and administrative aspects of air pollution.

Evaluate the legal and administrative practices related to air pollution control.

★ The Department of Physical Chemistry of The University of Cambridge is offering a summer school on "Fast Reactions," August 12–19, 1961. Further details may be obtained from The Secretary of The Summer School, Lensfield Road, Cambridge, England.



Why Philip Morris drains with Vulcathene® in new McComas Research Center



40 LABS COMPLETELY EQUIPPED WITH MAINTENANCE-FREE ACID-RESISTANT DRAINLINES

Plumbing Contractor: B. & G. Olsen Co., Richmond, Va.

This modern multi-million dollar research center is equipped with the newest in lightweight, non-corrosive drainage systems. Vulcathene meets *every* lab need because it's the most complete line of sinks, traps, pipe and fittings, in sizes from ½" to 6". Its smooth, non-adhering bore is scale-proof, clog-proof to chemical sludge. And, Vulcathene is an economical, quality system—assembled in seconds by the patented Polyfusion® method, or with leak-proof, hand-tightened couplings—at half the cost of other systems. It will pay you to find out more about Vulcathene.

NEW CATALOG tells all about this trouble-free, maintenance-free way to drain corrosive wastes. Write Dept. 342.

American Vulcathene

P.O. BOX 365 ROCHESTER 2, NEW YORK

DIVISION OF THE NALGE CO. INC

★ Establishment of the 1961 College Chemistry Teacher Awards program, honoring six outstanding college chemistry teachers throughout the United States, has been announced by the Manufacturing Chemists' Association. This is the fifth consecutive year in which the national chemical industry trade association has made these $1000 awards. Purpose of the awards is to honor the men and women engaged in college science teaching, call public attention to the importance of good science teaching at the undergraduate level, and inspire qualified students to choose careers in science teaching.

★ The National Science Foundation announces that March 1, 1961 is the next closing date for receipt of proposals in the Graduate Laboratory Development Program. Proposals received after March 1 will be reviewed following the next closing date, September 1, 1961. This program requires at least 50 percent participation by the institution with funds derived from non-Federal sources.

Purpose of the grants is to aid institutions of higher education in modernizing, renovating, or expanding graduate-level basic research laboratories used by staff members and graduate students. Only those departments having an ongoing graduate training program leading to the doctoral degree in science at the time of proposal submission are eligible.

Proposals, as well as requests for additional information, should be addressed to: Office of Institutional Programs, National Science Foundation, Washington 25, D. C.

★ A new method for obtaining uniformly sized fine particles to be pressed into KBr pellets is aiding infrared spectroscopists at Shell Oil Co's Houston Laboratory. Infrared spectra of the particles in the potassium bromide pellet show little or no scattered energy, since the particles are fine-ground to smaller dimensions than the wavelengths of the incident radiation.

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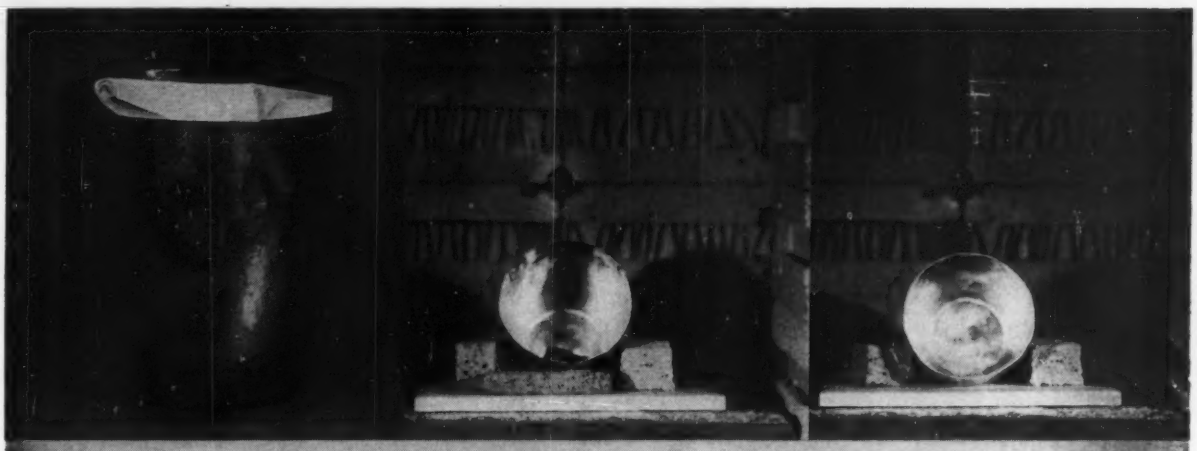

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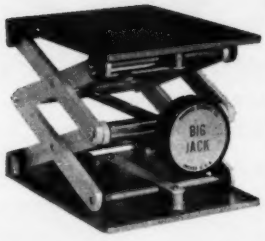

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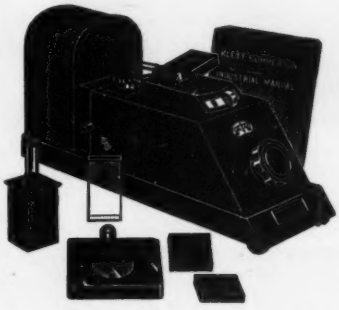

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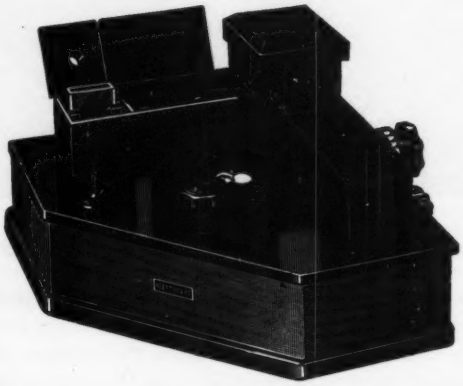

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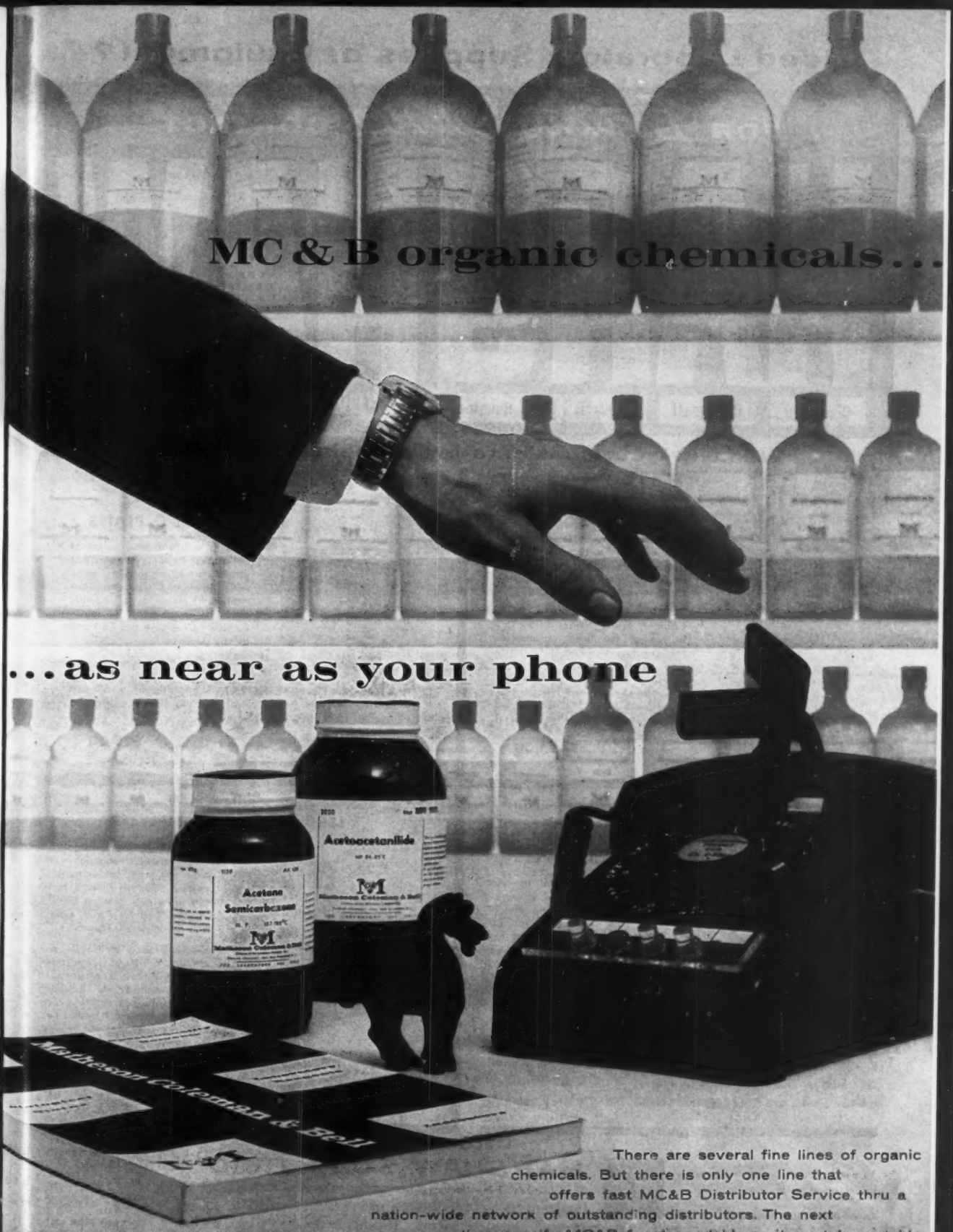

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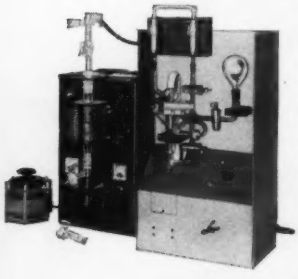

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THE EFFECT OF TEMPERATURE ON SOLUBILITY

A Chem Ed TESTED DEMONSTRATION

Rewritten from: Kenneth A. Kobe, THIS JOURNAL, **16,** 183 (1959) by F. B. Dutton
Checked by: Vance Hamilton, East Lansing High School, Michigan

PREPARATION

A saturated solution of manganese (II) sulfate is prepared at 27°C by shaking 70 g $MnSO_4$ or 130 g $MnSO_4 \cdot 7H_2O$ with 100 g water containing a few drops of H_2SO_4. The clear solution is decanted and stored in a stoppered test tube or preferably one which is sealed off and is consequently a "permanent" demonstration.

DEMONSTRATION

Pink crystals of $MnSO_4 \cdot 7H_2O$ separate out if the tube is placed in a mixture of salt and ice. Upon heating to 27°, complete solution takes place and on further heating above 27°, white $MnSO_4 \cdot H_2O$ separates out.

REMARKS

This demonstration serves to illustrate both the common increase of solubility with an increase in temperature and the less common decrease in solubility with increasing temperature (of a different hydrate) of the manganese salt. A solubility diagram is given in the original Kobe article. See also BATEMAN, L. A., and FERNELIUS, W. C., THIS JOURNAL, **14,** 315 (1937).

SURFACE TENSION

A Chem Ed TESTED DEMONSTRATION

Rewritten from: C. R. Caryl, THIS JOURNAL, **17,** 468 (1940) by F. B. Dutton
Checked by: Vance Hamilton, East Lansing High School, Michigan

PREPARATION

Provide two large beakers, one containing water, and the other a suitable wetting agent in appropriate concentration, such as a 0.1% Aerosol OT solution.

DEMONSTRATION

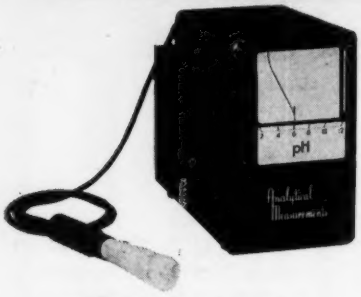
Powdered sulphur is poured from both hands into the beakers simultaneously. It is not wet by the pure water but pours like sand into the beaker of water containing the surface active agent.

REMARKS

Many variations will occur to demonstrators, including the use of string which may be lowered slowly onto the surface of the liquid in each beaker. It will accumulate on the top of the pure water but settle rapidly in the water containing the surface active agent. This is true also for balls of *non*-absorbant cotton and, of course, if one uses larger containers, for ducks.

Another variation is to add a drop of concentrated detergent solution to the beaker containing the floating sulfur. The spread of the detergent can be watched by the falling sulfur "snowstorm" which occurs.

pH At Your Fingertips



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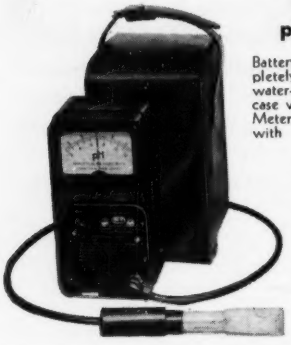

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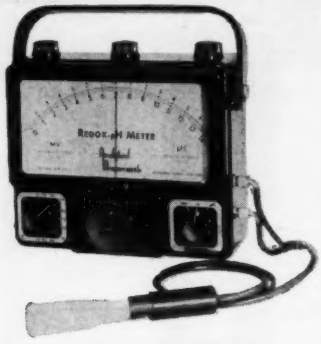

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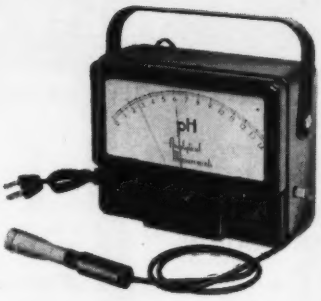

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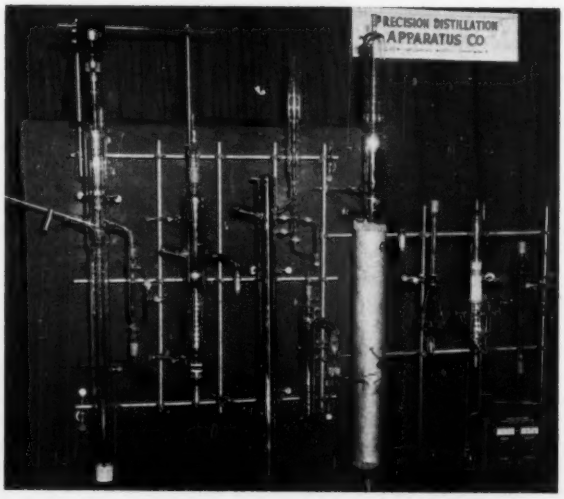

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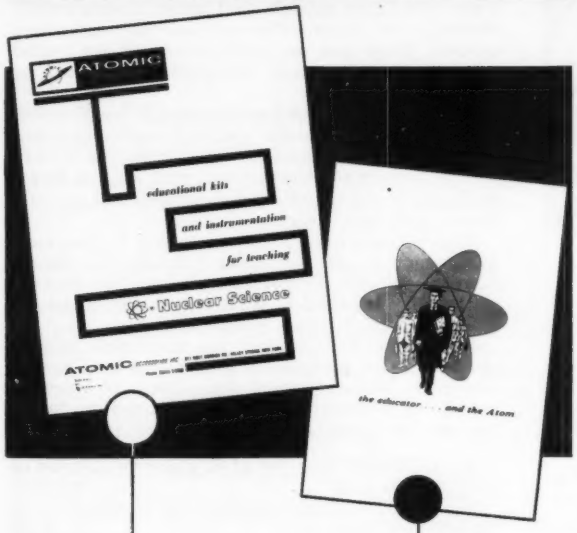

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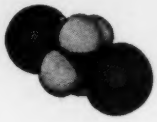
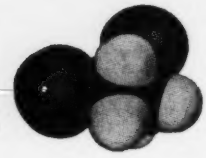
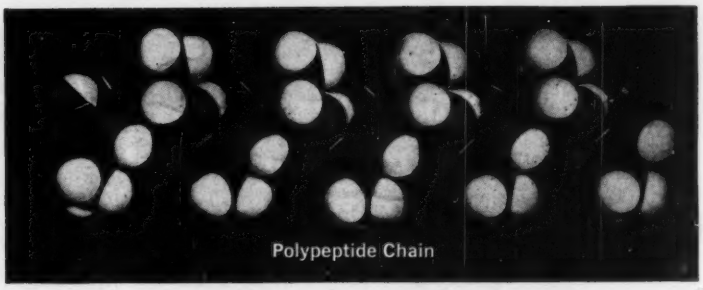
Dichloroethane



Cis- ("angular")
Dichloroethane





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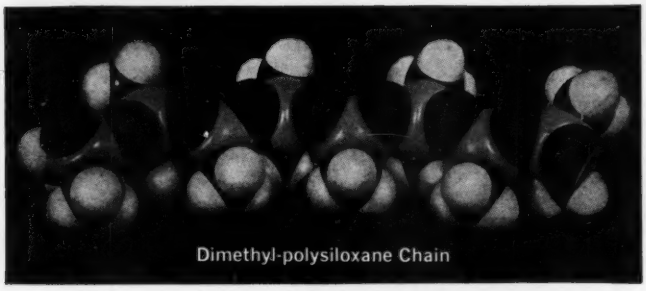

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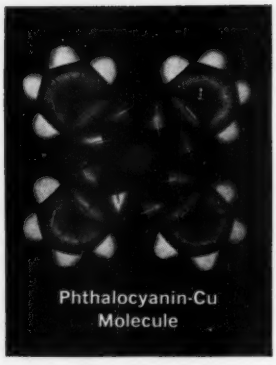
Phosphoric Acid



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CHEMICAL PROJECTS

Research Ideas for Young Chemists

Compiled by **JAY A. YOUNG,** Kings College, Wilkes-Barre, Penna.
JOHN K. TAYLOR, National Bureau of Standards, Washington, D. C.

Principles of the Systematic Separation of Cations

See WEISZ, H., *J. Chem. Educ.*, **33**, 334–5 (1956).

The chemical principles that determine the choice of procedure to be used in qualitative analytical separation and identification are discussed without reference to mathematics. Students who wish to undertake a project in qualitative analysis will find this article helpful as part of their preliminary study in preparation for the laboratory work to be performed. Many helpful hints to the answers to the questions cited in connection with other articles on this subject will be found in this paper.

Studies of Egyptian Bronzes

See WEISZ, H., *J. Chem. Educ.*, **32**, 70–2 (1955).

Ancient and valuable artifacts made of bronze are of interest to the chemist as well as to the antiquarian. The chemist is interested in the composition of these ancient alloys. The antiquarian is just as interested, though for different reasons, and is unwilling to allow destruction of the article to satisfy the chemist's curiosity. This impasse can be avoided by the Weisz ring-oven technique since it requires only a very small sample for analytical purposes, which can be obtained by rubbing a roughened glass microscope slide across the specimen several times. The streak of metal is dissolved in a drop of acid and the solution placed in the center of a filter paper disc. This drop is then exposed to hydrogen sulfide (two filtering flasks, one inverted over the other, can be used in place of the apparatus illustrated). Following this treatment, the paper is placed over the central hole of a metal ring. The ring is heated electrically while suitable reagents are added to the center of the filter paper. Dissolved ions are thereby transported across the paper and deposited on a circle immediately beyond the outer periphery of the heated ring. Successive "fractionations" readily yield identification of the constituents of the alloy.

Questions:

In terms of their composition, which trophies in your school trophy case were probably made by the same manufacturer? Or, in terms of composition and total weight, which trophies are the most valuable?

Is the composition of foreign silver (or copper) coins the same as that of domestic coins?

If available, determine the composition of similar coins of differing ages and identify any regularly varying trends in their composition.

Metal fixtures (plates covering electrical switches and outlets, faucets, etc.) installed in remodeled portions of older buildings are often different in style and in composition from the original fixtures. In the remodeling, defective fixtures in the older portions of the building are often replaced with new fixtures of the same type as those installed in the new, remodeled, portion. Older fixtures in good condition are not replaced. Can you determine that in a remodeled building available to you (permission should first be obtained) some metal fixtures in the unremodeled portions were defective at the time the remodeling was performed? Note: Many questions of historical interest can be answered by using the Weisz ring-oven technique. The two preceding questions are listed here, not so much to indicate a specific proposal for investigation, but to indicate the type of information that can be obtained. Local conditions will determine the nature of similar questions which will be more interesting to the chemist-detective (forensic chemist) who undertakes this kind of project.

Using Fluorescence in Qualitative Analysis

See WHITE, C. E., *J. Chem. Educ.*, **28**, 369–72 (1951).

Fluorimetric identifications are easily made and the apparatus required is inexpensively constructed from available components. Directions for the construction of the "visual comparator" and for fluorimetric identification of aluminum, zinc, boron, sodium, beryllium, thallium, and thorium are given. Other metals can also be identified; details can be found in the references cited by the author. Separative procedures are given for some of the metals listed; for others, separations must be devised by the investigator or found in other references.

Questions:

Determine the sensitivity of a fluorimetric identification procedure for one or more elements.

Some of the procedural steps immediately prior to the fluorimetric identification of an element are critical; slight deviations will have an adverse effect upon the identification of the element sought. From your laboratory investigation on any selected element, determine the reasons for the specific, detailed nature of these steps.

Raoult's Law Experiment for the General Chemistry Course

See HARRIS, F. E. AND NASH, L. K., *J. Chem. Educ.*, **32**, 575–7 (1955).

By the separate introduction of benzene, ethyl ether, and a mixture of these, into the air space in a partially filled graduate inverted in a beaker containing brine, data are obtained which permit a student to determine the validity of Raoult's Law. Other accessory data, such as the temperature of the gaseous mixture, the barometric pressure, etc., are also needed. The procedure is not complicated (though there are moderate fire and health hazards arising from the choice of liquids) and can be performed in a few hours. However, as indicated by Harris and Nash, there are several errors inherent in the method suggested. Students interested in Raoult's Law and its applications might wish to perform this experiment first, and then study the matter in a more precise way. The questions below probably cannot be answered by following the explicit procedure outlined by Harris and Nash.

Questions:

Using Raoult's Law as your basic principle, determine the molecular weight of a non-volatile solute dissolved in a volatile solvent by measuring the change in vapor pressure of the solvent when the solute is added at a convenient, fixed temperature.

Many pairs of liquids follow Raoult's Law rather closely, while other pairs show either a positive or a negative deviation from this principle. Select three pairs of liquid substances: one which you expect will follow Raoult's Law, one which will show a positive deviation, and one which will show a negative deviation. Determine whether your prediction is correct or incorrect in all three cases. Suggest reasons why your prediction was correct or incorrect and determine the validity of your reasons by further laboratory work.

New Vapor-Pressure Apparatus

See COLGATE, S. O. AND WHEALY, R. D., *J. Chem. Educ.*, **32**, 484–5 (1955).

Many devices have been designed to measure the vapor pressure of various substances (see annual and cumulative indexes of THIS JOURNAL). The apparatus described in the reference measures vapor pressure directly and is one of the simplest reported in the literature, requiring a minimum of glass blowing in its construction; it uses a vacuum pump and aspirator in its operation. The vapor pressure of a solid or a liquid can be measured with equal ease.

Questions:

Select any solid or liquid and measure its vapor pressure as a function of temperature. From the data obtained, calculate the heat of sublimation or vaporization of the substance selected.

Show that the vapor pressure of any pure substance is not proportional to its temperature. Suggest reasons why this might be true and test your reasons by further experimental work.

Is the vapor pressure of a super-cooled liquid greater than, equal to, or less than the vapor pressure of the solid form of the same substance at the same temperature? Explain, and confirm your explanation by further tests.

Measure the vapor pressure of a solution of salt dissolved in water, and of pure water, in the temperature range −10° to +10°C. Explain why salt will cause ice to melt when salt is sprinkled on ice in the winter.

Show that for any pure substance, the vapor pressure of the liquid form and of the solid form are the same at the melting point of the substance.

Is it true or false that the logarithm of the vapor pressure of a pure substance is a linear function of the reciprocal of the absolute temperature of that substance?

Select a solid, measure its vapor pressure as a function of temperature and, by any suitable means, measure its heat of fusion. Calculate the heat of vaporization of the substance selected, and compare this value with the value obtained by experimental means.

Is there a relation between the nature of the interatomic bonds or the intermolecular bonds in a compound and the vapor pressure of that compound?

Is the vapor pressure of a mixture of two liquids equal to, greater than, or less than the sum of the vapor pressures of the two pure liquids at the same temperature? Does the answer to this question depend upon the nature of the two liquids? If so, can you suggest an explanation?

From data which you obtain by vapor pressure measurements, calculate the composition of "constant boiling" hydrochloric acid. Confirm your calculation by preparing some constant boiling hydrochloric acid and measuring its concentration.

Show that, at ordinary pressures, it is impossible to obtain pure ethyl alcohol by fractional distillation of a mixture of water and alcohol.

Phase-Study Apparatus for Semi-Micro Use Above Atmospheric Pressure

See MARSHALL, W. L., WRIGHT, H. W., AND SECOY, C. H., *J. Chem. Educ.*, **31**, 34–6 (1954).

Small quantities of pure substances or mixtures are heated to temperatures of 300°C, or higher, in a sealed ampoule. Increase of pressure, though not measurable in the apparatus described, is usually insufficient to rupture the ampoule; with the precautions described, this is no more hazardous than any typical experimental investigation. The ampoule is placed in a cylindrical hole in the center of a cylinder of aluminum or graphite and the contents observed as the temperature is raised. Marshall *et al.* recommend that a continuous recorder be used to indicate the temperature; this is not necessary, though it would be a convenience.

Questions:

In what way does the behavior of a liquid-vapor meniscus depend, at the critical temperature, upon the degree of fractional filling of an ampoule with liquid, before the temperature is increased?

Determine the solubility of a series of related salts (such as lithium, sodium, potassium, and rubidium sulfates) as a function of (high) temperature.

Select any two substances which form immiscible liquids when melted, and draw a phase diagram for this system from the data you obtain.

What reagents can you find which will not react when heated in a test tube but will react when heated in an ampoule in the apparatus described? Identify the products of such a reaction, and suggest reasons why the reaction did not occur at ordinary pressures and slightly elevated temperatures. Test your reasons by further experimental work.

Is there any relationship between the critical temperature of a liquid and its normal boiling point?

From your analysis of the composition of a mineral or rock available in your vicinity, and from other observations, can you use the apparatus described by Marshall *et al.* to suggest a plausible description of the geologic history of the mineral or rock?

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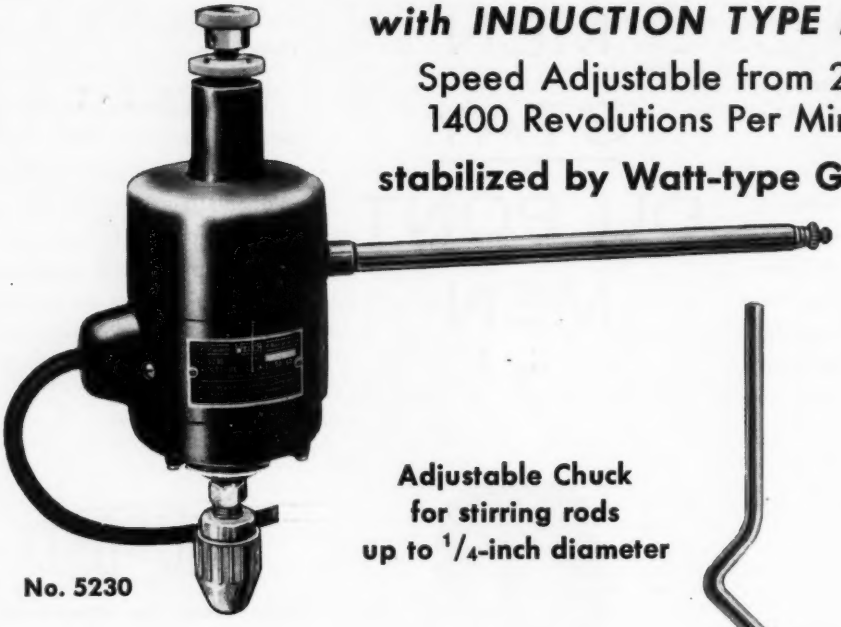

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Chem Ed

EDITORIALLY SPEAKING

An article in this issue which should receive careful and thoughtful reading by all chemists is Professor Guggenheim's "The Mole and Related Quantities." In simple unambiguous language it sets forth what is probably the primary unifying concept in the modern chemist's view of the materials with which he deals. Only the flimsiest semantic arguments can be found with the statement: *"The mole is the amount of substance containing the same number of molecules (or atoms or radicals or ions or electrons as the case may be) as there are atoms in twelve grams of carbon-12."*

Teachers of chemistry get their firmest feeling of success when students demonstrate that they know what "mole" means. When the student can do this not by writing a definition in so many words but by *using* the concept, he begins to think as a chemist does. The important recognition comes when the term "mole" stands for a definite *number* of any particular species. Textbooks have long lagged behind the prevailing pattern of thinking by the best teachers of chemistry on this point. One looks in vain for even the use of the word in all but the newest high school texts. The term is used in most texts for the introductory college course, but usually the student is left only with the idea that a "mole" is a "number of grams . . ."

Professor Guggenheim, later in his discussion, gives emphatic interpretation to his use of the word "amount." This is the implication which needs to be underscored for the neophyte chemist. Although perhaps unconsciously (as with some teachers, alas!), the working chemist now thinks in numbers of reacting species rather than in weights or even relative weights of materials involved. Certainly as soon as he anticipates the measurement of energy associated with a given reaction he must consider Avogadro's number of events at the atomic level as a standard for reference.

A second implication grows out of this *number* definition. This is the inclusion of the parenthesis in Professor Guggenheim's statement. Here again, common usage by working chemists transcends the textbook definition by not limiting the mole concept to gram-molecular weight. A student who realizes that the Faraday represents the charge of a mole of electrons will have at his disposal the key concept to applications ranging from electrolysis to redox titrations.

Another big advantage is built into this definition which emphasizes number of species: it is a concept which can grow in significance as a student's chemical knowledge accumulates. It may be expedient to base the first use on the weight idea, for example, in the stoichiometry which may appear very early in an elementary course. Later it will not be tossed aside nor even strained to encompass more sophisticated theory. We recall once receiving some letters (we keep them in an asbestos file) castigating us for publishing an article which defined a mole as a weight containing 6.023×10^{23} molecules and then applied that definition to an amount of HNO_3, S, or $Bi(NO_3)_3$. The point of the article was to emphasize the advantage of the mole concept in the first introduction of a high school student to stoichiometry. We admit to the editorial sin of laxity in not using the blue pencil to introduce the term "formula weight." However, we doubt that there would be any ambiguity in the mind of a student at that early stage of his first course. He was being taught to make proper *use* of the concept to solve stoichiometric problems based on a complicated but balanced equation describing unfamiliar chemistry. We doubt, too, that this initial working concept of mole could block the student's later appreciation of what modern theories of electrolytes or bonding in solid substances could add to his earliest recollection (that coefficients in a balanced equation establish the relative numbers of reacting moles). We doubt that a teacher would have trouble later in convincing him that a *mole* of Bi atoms will lose three *moles* of electrons to form a *mole* of Bi^{+++} ions which probably will react in aqueous solution to form a variety of other species whose *molar* concentration may be calculated if appropriate equilibrium constants are known, etc.

There is an interesting contrast between the modern problem presented by this article and Dr. Ihde's account of the circumstances under which the Karlsruhe Conference was convened by Kekulé a century ago. Chemistry, still a young science, has moved far in 100 years. Yet then as now, the magnificent strength of a unifying concept was the basis for the advance of chemistry. Then it was a gradual resolving of chaos; now it is the correlating and consequent simplifying of diverse phenomena. Cannizzaro's advocacy of Avogadro's hypothesis was based on his use of it ". . . to lead my students to the conviction which I have reached myself." We doubt if anything more important than a unifying concept has been invented in 100 years of successful chemistry teaching.

A ChemEd SYMPOSIUM

Cosmochemistry

Introduction

Chemistry and geology have had close connections since the beginning of each science, yet their daughter geochemistry has been recognized as a separate science only since the turn of the present century. In the same way, the chemical aspects of the extraterrestrial universe were being investigated by chemists, astronomers, and physicists for a long time before recognition of the distinct discipline of cosmochemistry. This recognition followed the intense revival of academic research after World War II, and was crystallized by the appearance of the journal *Geochimica et Cosmochimica Acta* in 1950.

It was appropriate, therefore, that at a meeting of the grandmother society there should be a symposium on the daughter science, and that the symposium should include a session on the granddaughter field. At the 1960 Spring Meeting of the ACS at Cleveland, a "Symposium on Geochemistry: Analysis and Synthesis" was sponsored jointly by the Divisions of Inorganic Chemistry, Physical Chemistry, and Chemical Education. The Symposium was organized by Professor Rustum Roy of Pennsylvania State University, and the session on Cosmochemistry was arranged with the assistance of the present writer.

This session featured the Acceptance Address of the recipient of the Nuclear Applications in Chemistry Award, Professor Charles D. Coryell of MIT. This prize, sponsored for the sixth consecutive year by the Nuclear-Chicago Corporation, carried the citation:

> . . . .for researches on the characterization of the products of nuclear fission including the new element promethium; for development of methods for making the byproducts of atomic energy into useful tools of science; for applications of radioisotopes in chemistry; for inspiration of countless students of nuclear chemistry; for scientific service to his country and devotion to the orientation of nuclear science toward peace.

Professor Coryell's paper described the latest ideas concerning one of the oldest scientific questions posed by man. The importance of nuclear chemistry, and particularly of an understanding of the fission process and products, in unravelling the more complicated processes and products of nucleosynthesis was clearly evident.

One of the critical sets of empirical data on which theories of element formation are based was discussed by Professor William D. Ehmann of the University of Kentucky. He mentioned his own and others' application of nuclear techniques to meteorite analysis.

Professor Edward Anders of the University of Chicago presented a paper prepared jointly with Mr. Gordon G. Goles. He showed how applications of physical, inorganic, and nuclear chemistry to the study of meteorites has shed much new light on the history of these bodies and indeed on the early history of the solar system as a whole.

The final paper in the session discussed how the recent first observation of an extinct natural radionuclide and theoretical developments in astrophysics have made possible calculations of the time interval in which our elements were formed and of the subsequent generation of radioactive heat, and have presaged further observations of manifestations of extinct natural radioactivity.

The large audience and lively discussions at the Cosmochemistry session attested to the rapid growth of interest in this field. Whether it will assume the status of a full-fledged scientific discipline—with a substantial body of scientists referring to themselves primarily as cosmochemists—remains to be seen.

Truman P. Kohman, *Chairman of Symposium*
Carnegie Institute of Technology, Pittsburgh, Pennsylvania

EDITOR'S NOTE: The award of the Newcomb Cleveland Prize by the American Association for the Advancement of Science to Professor Edward Anders lends further distinction to the publication of this symposium. At the 127th meeting in New York, December 1960, the AAAS presented this one of its four major awards for "a noteworthy paper, representing an outstanding contribution to science," selected from those read at last year's AAAS meeting. The paper included here is not the award address, but discusses the same general theme of the author's investigations.

—The cover

The chief process by which heavy nuclei synthesized in stellar interiors are distributed throughout galactic space is dramatically evident in this photograph of the Crab Nebula. Technically known as Messier 1—the first object listed in the French comet-hunter's catalog of 103 nebulae and star clusters published in 1784—it has been identified with a "new star" recorded by Chinese and Japanese astronomers in 1054 AD and with the third brightest discreet radio source in the sky, Taurus A. It is presumably emitting cosmic radiation, which has not yet reached us because of galactic magnetic fields. Its mass is estimated as about 15 times that of the sun, and at the center is a faint super-hot super-dense "white dwarf" star, the slowly dying remnant of a once-violent supernova. The turbulent gases it ejected are now expanding at a rate of about 1400 km sec^{-1} and have already attained a radius of about 4 light-years, about the distance from the sun to its nearest stellar neighbor. It is about 5000 light-years distant, and visible as a faint "planetary nebula" in a small telescope. This photograph in the red light of the hydrogen Hα line was made with 100-in. Hooker telescope, Mt. Wilson observatory. Photograph: courtesy of California Institute of Technology.

William D. Ehmann
University of Kentucky
Lexington

Recent Improvement in Our Knowledge of Cosmic Abundances

A knowledge of the relative abundances of the chemical elements on a cosmic scale is prerequisite to the development of a theory for the synthesis of the elements in the stars. Since our sun is considered to be a relatively "average" star, we can as a first approximation equate the terms "cosmic abundances" and "solar system abundances." However, determination of these solar system abundances has still been a formidable problem.

Much of the early abundance data was derived from geochemical investigations of the crust, oceans, and atmosphere of the earth. More recently, significant contributions to this field have been made by spectrographic investigations of the light emitted by the sun and the stars (*1*) and by chemical analyses of the meteorites. Some information on the distribution of hydrogen in the universe has been obtained recently through the use of radiotelescopes.

It was only natural to look to the meteorites for cosmic abundance data, since they represent the only extra-terrestrial specimens available for laboratory analysis. It has been suggested by Urey and Craig (*2*) that a certain class of stony meteorites (aerolites) called chondrites might represent the best average sample obtainable for the determination of the relative abundances of the non-volatile elements in our solar system (Fig. 1). In general, meteoritic matter appears to be fractionated into three major phases: the metal phase, the sulfide phase, and the silicate phase. The per cent by weight of each of these phases in meteoritic matter is estimated to be approximately 10%, 5%, and 85%, respectively. Certain of the elements are known to concentrate predominantly in one or the other of these phases. Therefore, it is possible to analyze a single meteorite phase in which we are quite certain the element in question is concentrated, and using the relative ratios of the various phases as stated above, compute an elemental abundance for all meteoritic matter. This has been done particularly for the determination of the abundances of some of the more noble metals that are presumed to be concentrated in the metallic phase. Analyses of the iron meteorites (siderites) for these elements have been used in some cases to compute cosmic abundance data.

The difficulty with this procedure is that there is still a measure of uncertainty as to the true values of these phase ratios and the true distribution of the elements between them. Analysis of the chondrites which contain all three meteoritic phases appears to be the most reliable approach to the determination of elemental abundance data at present, and will be stressed in this paper. Largely on the basis of their structure, Urey postulated that the chondrites are made up of rubble-like material, both metallic and non-metallic, which resulted from collisions between asteroid-sized bodies early in the history of the solar system. This rubble aggregated into layers on the larger asteroidal bodies of approximately lunar size. Finally, at some later time, a collision between two of the larger bodies occurred, resulting in the compression of the layers of rubble into the objects known as chondrites. More recently, Urey (*3*) has suggested a slightly modified collision theory in which the moon may have had a role in chondrite formation.

Anders and co-workers in this symposium (*4, 5*) depart sharply from the suggestion that a collision mechanism is necessary to explain chondrite structure. They have proposed that a unique type of volcanic action on asteroid-sized bodies may explain many aspects of chondritic structure.

Regardless of which of these theories proves to be correct, Urey's original suggestion that the chondrites are representative of the non-volatile elemental abundances in the solar system is a useful starting point for the determination of cosmic abundances until that time when better samples become available through extra-terrestrial exploration.

Suess and Urey (*6*) in 1956 compiled a table of cosmic abundances based on various types of meteoritic, solar, and terrestrial elemental abundance data. In addition, the accurate relative isotopic abundances available from mass spectrographic analyses were used to derive isotopic abundances in the Suess and Urey table. A plot of the data from these tables shows a significant

Presented as part of the Symposium, "Geochemistry: Analysis and Synthesis," before the Divisions of Inorganic Chemistry, Physical Chemistry, and Chemical Education at the 137th Meeting of the ACS, Cleveland, Ohio, April 1960.

This work was supported in part by the U. S. Atomic Energy Commission.

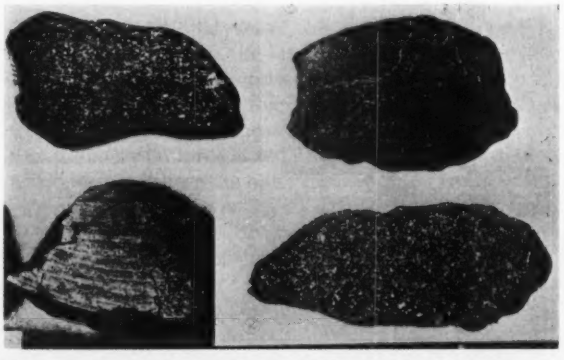

Figure 1. Cross-sections of four chondritic meteorites illustrating their unique structure; from Nininger (*44*).

amount of "fine structure" (Fig. 2). The current theories that describe the synthesis of the elements in stars have drawn heavily from this extensive compilation of abundance data. Detailed mechanisms for element synthesis have been given most recently by Burbidge, *et al.* (*7*) and Cameron (*8*, *9*). Coryell (*10*) in this symposium has presented some recent information relating to these theories.

In this paper only the experimental abundance data that have appeared since the original Suess and Urey paper will be discussed. For the most part, this paper will deal with new data resulting from refined techniques of analysis, as applied to the chondritic meteorites. Some implications of this new data with regard to the present theories of nucleosynthesis and areas for future research will be mentioned.

Recent Abundance Data

In their 1956 paper Suess and Urey (*6*) pointed out the need for better analytical data for many elements. Indeed, in some cases no analytical data at all existed for certain of the elements in chondrites, and cosmic abundance values were assigned by interpolation.

During the past several years, widespread use has been made of a number of new analytical techniques, such as isotope dilution analysis and neutron activation analysis. These techniques have been applied with considerable success to the determination of elemental and isotopic abundances in meteorites. The data for chondrites in Table 1 express the non-volatile elemental abundances as atomic abundances relative to silicon equal to 10^6. The silicon is taken to represent 18.5% by weight of primitive solar non-volatile material. These new data are compared to the corresponding data from the compilation of Suess and Urey (*6*).

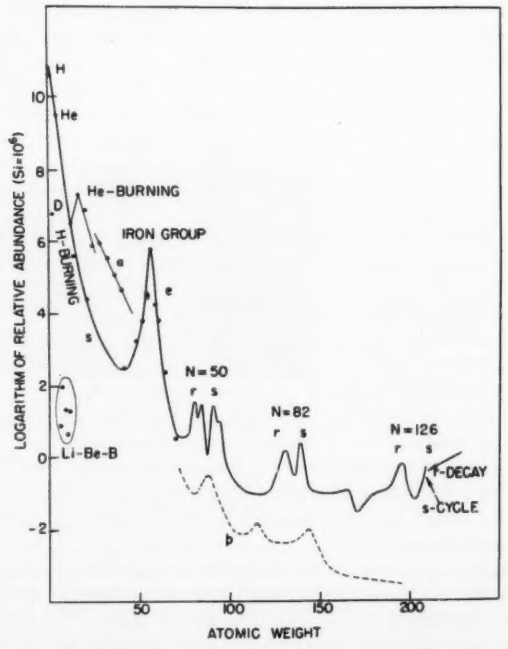

Figure 2. Schematic curve of atomic abundances as a function of atomic weight based on the data of Suess and Urey (6). Small letters refer to the types of nuclear processes resulting in element formation in a given region; from Burbidge, Burbidge, Fowler and Hoyle (7).

The number of specimens refers to the number of different meteorites used in obtaining the average abundance value listed. The simple average of the average abundance in each of the meteorites analyzed is used for the most part in this table. With small numbers of samples, different methods of averaging data may lead to small variations in the numerical average. Data for a few achondrites are included in the averages, where their abundances were found to be virtually the same as for the chondrites. In cases where significant differences in the abundance data for chondrites and achondrites were observed, Table 1

Table 1. Atomic Abundances from Recent Chondrite Analyses

Z	Element	Ref.	Number of specimens	Atomic abundance (Si = 10^6)	Suess-Urey (Si = 10^6)
3	Li	(*11*)	3	38	100
19	K	(*12*)	3	3,290	3,160
20	Ca	(*13*)	7	45,000	49,000
21	Sc	(*14*)	5	32	28
22	Ti	(*15*)	43 Falls	2,090	2,440
		(*15*)	45 Finds	1,900	....
24	Cr	(*14*)	5	6,400	7,800
25	Mn	(*15*)	43 Falls	7,200	6,850
		(*15*)	45 Finds	6,400	....
27	Co	(*16*)	8	1,190	1,800
28	Ni	(*16*)	8	24,800	27,400
29	Cu	(*16*)	7	186	212
32	Ge	(*17*)	8	18.7	50.5
34	Se	(*18*)	4	18.8	67.6
37	Rb	(*19*)	12	4.6	6.5
		(*12*)	5	5.8	....
44	Ru	(*20*)	4	(1.2)[a]	1.49
45	Rh	(*21*)	5	0.27	0.214
47	Ag	(*21*)	5	0.131	0.26
49	In	(*21*)	5	0.0013	0.11
52	Te	(*18*)	4	0.73	4.67
		(*24*)	7	(0.50–4.0)[a]	....
53	I	(*24*)	8	(0.044–0.67)[a]	0.80
55	Cs	(*19*)	12	0.10	0.456
		(*12*)	5	0.14	....
56	Ba	(*22*)	4	3.96	3.66
		(*15*)	43 Falls	6.44	....
		(*15*)	45 Finds	76.2	....
57	La	(*23*)	2	0.40	2.00
58	Ce	(*23*)	2	0.62	2.26
59	Pr	(*23*)	2	0.15	0.40
60	Nd	(*23*)	2	0.74	1.44
62	Sm	(*23*)	2	0.25	0.664
63	Eu	(*14*)	5	0.078	0.187
		(*23*)	2	0.097	....
64	Gd	(*23*)	2	0.36	0.684
65	Tb	(*23*)	2	0.056	0.0956
66	Dy	(*23*)	2	0.39	0.556
67	Ho	(*23*)	2	0.078	0.118
68	Er	(*23*)	2	0.21	0.316
69	Tm	(*23*)	2	0.039	0.0318
70	Yb	(*23*)	2	0.19	0.220
71	Lu	(*23*)	2	0.036	0.050
73	Ta	(*48*)	8	0.019	0.065
74	W	(*45*)	8	(0.10)[a]	0.49
		(*48*)	5	0.11	....
76	Os	(*20*)	6	(0.6)[a]	1.00
77	Ir	(*46*)	6	0.38	0.821
79	Au	(*25*)	4	0.13	0.145
80	Hg	(*26*)	5	(0.076)[a]	0.284
81	Tl	(*26*)	5	0.0010	0.108
		(*27*)	5	0.00038	....
82	Pb	(*27*)	4	0.05–0.28	0.47
		(*28*)	3	0.15	....
83	Bi	(*26*)	6	0.0016	0.144
		(*27*)	5	≤0.005	....
90	Th	(*29*)	5	0.026	....
92	U	(*22*)	4	0.0072	....
		(*30*, *31*)	6	0.0079	....

[a] Numbers in parentheses are preliminary numbers from work still in progress.

lists only the chondrite data. The numbers in parentheses are tentative determinations based on work now in progress. These numbers may be subject to considerable revision upon completion of the experimental work, but are included for completeness. A small number of analyses limited to a single meteorite, or to the troilite phase of meteorites, were not included in this tabulation.

Table 2 lists analytical data on iron and stony-iron meteorites that have appeared since the Suess and Urey paper. Due to the previously discussed difficulties in the interpretation of data from a single meteoritic phase, these abundances are here listed merely in units of ppm by weight for these specific meteorites. In analyses for several of the elements listed in Tables 1 and 2, a wide range of values was obtained for different meteorite specimens. In these cases, where also the number of specimens analyzed was small, listing an average value might be misleading: Therefore, only the limits for the range of these experimental values are entered.

It is of interest to note that the majority of the results listed in Tables 1 and 2 were obtained by use of neutron activation analysis. The great sensitivity of this technique coupled with its inherent freedom from laboratory contamination makes it highly useful for determination of certain trace element abundances.

Table 2. Recent Analyses of Iron and Stony-Iron Meteorites

Z	Element	Ref.	Number of specimens	Parts per million by weight
24	Cr	(*32*)	17	<5–185
		(*33*)	88	37
27	Co	(*33*)	88	5,100
28	Ni	(*34*)	8	74,200
29	Cu	(*32*)	17	7.8–434
		(*33*)	88	152
31	Ga	(*33*)	88	34
32	Ge	(*32*)	17	0.2–359
		(*33*)	88	107
33	As	(*32*)	17	0.6–33
57	Sb	(*32*)	17	0.01–0.78
73	Ta	(*48*)	3	0.0026
74	W	(*48*)	3	1.67
75	Re	(*35*)	10	0.224
76	Os	(*35*)	10	2.03
90	Th	(*36*)	2	6×10^{-6} to 2×10^{-5}
92	U	(*37*)	8	10^{-6} to 10^{-4}

Discussion

The response to the plea of Suess and Urey (*6*) for more and better analyses of chondrites is quite remarkable, as indicated by the length of Table 1. It is necessary, however, to be quite cautious about substituting some of the new data for the Suess and Urey values. It is to be noted that most of the results in Table 1 are based on the analyses of ten or less meteorite specimens, while in some cases the older data resulted from a much larger number of analyses. In addition, the recent work of Moore and Brown (*38*) indicates that compositional differences may exist between chondrite "falls" and chondrite "finds." These differences for barium are approximately an order of magnitude.

Therefore, in this paper no attempt will be made to reconstruct the Suess and Urey cosmic abundance curve, using the new analytical data, although certain of the data undoubtedly represent the best available for a given element. Until additional analytical data are available and the compositional differences between "falls" and "finds" further investigated, more definitive assignment of cosmic abundances from some of these data would be premature.

Li (Z = 3) to Cu (Z = 29)

With the exception of the new analysis for lithium which is based on only three chondrites, the new determinations listed in Table 1 are in reasonably good agreement with the Suess and Urey values in this region.

The investigation of the chondritic abundances of scandium and chromium was prompted mainly by rather large discrepancies between the Suess-Urey values based on older chondrite analyses and the solar spectra data of Aller (*1*). The spectral abundances for scandium and chromium relative to silicon = 10^6 are 40 and 13,000 respectively. It is seen in Table 1 that the new analytical data of Bate, *et al.* (*14*) tend to confirm the Suess and Urey values. Due to considerable uncertainties in the determination of the solar abundance for scandium, it would appear that its cosmic abundance is best approximated by the new chondritic data. For chromium the solar value has been more precisely determined, and may represent a truly high abundance in the sun's atmosphere. Alterations of relative concentrations of elements in the sun's atmosphere by mixing with the interior are possible and lead one again to rely more heavily on the chondrite analyses for the chromium cosmic abundance. It is to be noted that the newly determined abundances of titanium and manganese (*15*) are much more nearly the same for chondrite "falls" and "finds," than was the case for barium. This may mean that compositional differences between "falls" and "finds" are small for certain elements; hence, much existing data are adequate for computing cosmic abundances.

Ge (Z = 32) to Lu (Z = 71)

In this region some serious discrepancies are noted between certain of the new data and the Suess and Urey values. It has been suggested that neutron capture on a fast time scale produces abundance peaks at approximately $Z = 35$ (Br), $Z = 54$ (Xe), and $Z = 78$ (Pt). Since abundance data for bromine and xenon in chondrites would be unreliable due to their volatility, accurate abundance data on neighboring relatively non-volatile elements, such as $Z = 34$ (Se) and $Z = 52$ (Te) would be valuable in assessing the magnitude of these abundance peaks in this weight region.

Little analytical data existed for these elements before 1956. The abundance value for tellurium used by Suess and Urey was an interpolated value. The new analytical data for these elements and other neighboring elements, such as iodine, rubidium, and cesium, all appear to be lower than the Suess and Urey abundances. A wide range of abundances were obtained for iodine and tellurium, and the simple average would perhaps not be representative. Goles (*24*) has noted that the "ordinary" chondrites, such as used in most abundance determinations, have iodine and tellurium abundances near the low end of the range of values listed in Table 1. Higher abundances for these elements are found in both carbonaceous chondrites and enstatite chondrites. Therefore, assignment

of cosmic abundance values for these elements must depend on future research to determine if certain of these subtypes of chondrites might be more representative samples for cosmic abundance determinations than the "ordinary" chondrites. Due to the large difference between the barium abundances in chondrite "falls" and "finds" (*15*, *38*), it is difficult to assign a cosmic abundance value to barium at present. However, in contrast to the other new determinations in the xenon peak region, all the new data point to a barium abundance higher than that of Suess and Urey. It appears that a final evaluation of the magnitude of these fast time scale neutron capture peaks will require new determinations of other elements in these regions, such as arsenic, strontium, and antimony.

The abundance of iodine is of interest for an additional reason. Reynolds (*39*) has recently found anomalous amounts of Xe^{129} in the Richardton chondrite. This presumably results from the decay of I^{129} ($H = 2.5 \times 10^7$ years) which was trapped in the original parent body of the meteorite. These results imply that a period of approximately 3.5×10^8 years intervened between the end of nucleosynthesis and the condensation of the parent meteorite material to a point at which rare gases were retained. Wasserburg, Fowler, and Hoyle (*40*) interpret this number as a maximum value for the time interval. Assuming nucleogenesis occurred at a uniform rate over a time large as compared to the mean life of I^{129}, they calculate an interval of 2×10^8 years, using the data of Reynolds. These calculations involve the chondritic abundance of I^{127}, which Reynolds estimated to be approximately 1 ppm. Although this time interval is not extremely sensitive to the I^{127} abundance, the new iodine data for "ordinary" chondrites listed in Table 1 indicate that corrections in the calculations will undoubtedly be necessary.

The new value for the indium abundance (*21*) is approximately 100 times lower than the Suess and Urey selected abundance. It appears impossible to resolve this extremely low abundance and the low abundances of certain heavy elements such as mercury, thallium, lead, and bismuth, with the existing theories of nucleosynthesis. It may be that these extremely low abundance values in chondrites are due to some as yet not understood fractionation process that occurred prior to or during the formation of the solar system, including the parent meteoritic bodies. Fish, *et al.* (*5*) suggest that their quasi-volcanic recycle mechanism for the formation of chondrites would result in the depletion of such chalcophilic elements as indium, mercury, thallium, lead, and bismuth in the chondrites. Additional analytical data on various meteoritic phases are needed to explain these discordant abundances.

The new data on the rare earths, although based on only two chondrites, indicate slightly lower values than given by Suess and Urey. A detailed discussion of the significance of these data will not be attempted here. It is of interest to note, however, that the new values for the abundance of europium (approximately 0.08, $Si = 10^6$) are much closer to the Suess and Urey value of 0.187 than to the solar spectra value of 0.6 (*1*). As in the case of scandium, the solar value is subject to considerable experimental uncertainties and the best value for the cosmic abundance of europium is certainly derived from the new data of Table 1.

W (Z = 74) to U (Z = 92)

As mentioned in the previous section, the elements mercury, thallium, lead, and bismuth are all in much lower abundances in the chondrites than can be explained by the current theories for element synthesis. Since these are all chalcophilic elements, the theory of Fish, *et al.*, (*5*) offers the best explanation of these low abundances to date. The abundances of lead, thorium, and uranium have considerable significance in the field of cosmochronology. Details of these applications are discussed most recently by Kohman (*41*), in this symposium, and by Fowler and Hoyle (*42*).

Of great importance to these calculations is the present number ratio, Th^{232}/U^{238} in the chondrites. Bate, *et al.*, (*29*) listed a value of 3.6 for this ratio in chondrites based on their experimental results for thorium in five chondrites and the uranium results of Hamaguchi, *et al.*, (*22*) in four chondrites.

Recently some new data for uranium in six chondrites has appeared (*30*, *31*). Using these data for uranium and the Bate, *et al.*, data for thorium, the ratio Th^{232}/U^{238} becomes approximately 3.3. It would appear that the best approximation of this ratio that can be made at present would be close to the average of the previously mentioned two values, or 3.45. Slightly different values may be obtained, depending on how this average is taken. Additional experimental determinations of this ratio would be extremely valuable.

It has been pointed out by Hamaguchi, Reed, and Turkevich (*22*) and Marshall and Hess (*28*) that the amount of uranium and thorium found in chondrites is not consistent with the published isotopic composition of the lead in chondrites. In fact, the new abundance data for uranium and thorium are approximately 1.5 to 3 times too low. Here again, the explanation could well be loss of these elements by chemical fractionation processes that are not fully understood at present.

Summary

The analysis of meteorites has without question provided a great deal of information that is useful in the formulation of theories for nucleosynthesis. Although much important meteoritic abundance data have been obtained since the need of such data was pointed out by Suess and Urey (*6*), precise analyses for many elements are still lacking. Among the most important areas for future work are the following:

(1) It would be desirable to obtain additional new experimental data for the abundances of the elements in the region of the abundance peaks for neutron capture on a fast time scale. This data would allow determination of the magnitude of the contribution of this process in the regions of the bromine, xenon, and platinum abundance peaks. Experimental work is underway at the University of Kentucky to determine the abundances of tantalum, tungsten, iridium, and mercury in the region of the platinum abundance peak by use of neutron activation analysis. As noted in Table 1, new experimental work on osmium is underway at Argonne National Laboratory, using this same technique. In all these cases previous experimental data are meager or lacking.

(2) As discussed in this text, the ratio Th^{232}/U^{238} is highly important to the calculations involved in the field of cosmochronology. Additional abundance data

on these two elements would serve to pin down more closely the value of this ratio.

(3) Data on the abundances of the chalcophilic elements in each of the three principal meteoritic phases is needed in order to evaluate the theory of Fish, *et al.* (*5*).

(4) New data on the abundance of iodine from experimental work now underway (*24*) will enable more accurate calculations of the time interval between the end of nucleosynthesis and the condensation of the parent meteoritic bodies to the point of entrapment of the rare gases.

It is evident that cosmic abundance data obtained from the meteorites are based on the thesis that the chondrites are representative samples of the non-volatile matter in the solar system. The large discrepancies between some of the new meteoritic data and the abundances reasonably predicted on the basis of current theories of nucleosynthesis suggest that the "ordinary" chondrites may not be representative samples for determining abundances of certain elements. The possibility of chemical fractionation processes operating prior to, or during, the condensation of the parent meteoritic bodies must be considered as a possibility in the explanation of certain of these data. As the difficulties in the interpretation of solar and stellar spectra are resolved, some of the problems of translating chondritic into cosmic abundances may be eliminated.

A recent paper by Urey (*43*) discusses again the significance of the meteoritic elemental abundances. In this paper is included the statement, "We should not take meteoritic abundances as more than an approximation to true primitive values." In view of some of the new analytical data, this statement is well justified.

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Note Added in Proof

New analytical data for selenium and tellurium (*47*) and for barium, mercury, thallium, lead, bismuth, and uranium (*49*) have appeared in print, since the compilation of Table 1. In general, these data are in reasonably good agreement with the values listed in this paper. It is noted by Reed, *et al.* (*49*) that the abundances of the chalcophilic elements are much higher in the carbonaceous chondrites and enstatite chondrites than in the "ordinary" chondrites, which are commonly used for cosmic abundance work. Indeed, these higher abundances are in quite good agreement with values predicted on the basis of theories of nucleosynthesis. It is suggested that the carbonaceous chondrites may be more representative samples for the determination of cosmic abundances, at least for certain of the elements, than the "ordinary" chondrites previously used.

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Theories on the Origin of Meteorites

It is generally agreed that meteorites are fragments of larger bodies. There are, however, profound differences of opinion on the properties and dimensions of these parent bodies. A central point in this controversy is the nature of the processes that led to the structural and compositional features of the meteorites.

The chemist may have much to say concerning these problems. Some of the most crucial points, such as the dating of events in meteoritic history by the use of radioactive decay (*1*, *2*), and the investigation of elemental abundances to infer both the primordial composition of the solar system (*3*, *4*) and its chemical history (*5*, *6*), are best treated by chemical techniques. Chemical insight is also valuable in interpreting many of the observations on the detailed structure of meteorites. The laws of physical chemistry, derived from observations on the Earth's surface, may be fruitfully applied to treat events occurring in the outer reaches of the solar system as long as 4.5 aeons[1] ago.

It is important to keep in mind, however, that these measurements and observations on the meteorites relate to a sample which is only a minute fraction of the hypothetical parent body—perhaps 10^{-15} to 10^{-20}. Furthermore, it seems likely that this sample is biased in favor of material which would not easily be destroyed during the break-up of the parent body and which could survive passage through the atmosphere. Possibly the sample also is weighted in favor of material which was near the surface of the parent body. Any conclusions about the nature of the parent bodies drawn from experiments on this sample necessarily represent a long extrapolation.

Theories of the Origin of Meteorites

In one respect, all the attempts at understanding the origin of the meteorites begin on common ground: it is now generally believed that the accumulation of the parent bodies from the solar nebula occurred at temperatures below 600°K (*8*, *9*), and the chemical nature of this starting material has been investigated in some detail (*10*, *11*). The chief differences in the three principal theories of meteorite origin lie in the dimensions proposed for the parent bodies. These are postulated to have been (1) of planetary size; (2) two successive generations, one of lunar size and one of asteroidal size; and (3) of asteroidal size *only*. All of these theories have certain advantages and disadvantages in their application to explaining the observations on meteorites. It is probably best to regard them all as working hypotheses, subject to further experimental tests.

Planetary-Sized Parent Body

A number of investigators, among whom Ringwood (*12*) and Lovering (*13*, *14*) may be cited as recent examples, have expressed the view that the meteorites evolved in a single body, of a size intermediate between that of the moon and that of the earth. The chief advantages of this hypothesis are that it is apparently simple, with only one or at most two parent bodies required; that many analogies with the presumed composition and structure of the earth's interior can be drawn; and that the occurrence of diamonds in meteorites may be attributed to the graphite-to-diamond transformation at high gravitational pressures. Also, a source of heat for meteorite synthesis is available in the long-lived radioactivities which now heat the earth's interior, and would certainly have been more intense heat sources some four-and-a-half aeons ago.

There are, however, very serious disadvantages connected with these points. Breaking up a body of planetary size is extremely difficult and no one has proposed a truly satisfactory solution to the problem. It is necessary to suggest a plausible way to perform this disruption so as to eject materials from as deep as the core (which is postulated to be the place of origin of the iron meteorites) and at the same time to avoid the vaporization or profound metamorphosis of most of the ejected materials, both silicates and metal. One should note that the energy released in allowing two planets to fall together would be enormous, more than enough to vaporize them both if efficiently converted to heat (*15*). The mean energy per gram in the collision of two objects of equal radius a and equal mean density ρ is on the order of:

$$\langle E \rangle = \frac{4}{5}\pi G\rho(\sqrt[3]{2}\cdot a)^2 \qquad (1)$$

where G is the gravitational constant. If each were the size of the moon, with a density of 3.3 g/cm³, $\langle E \rangle$ would be ~640 cal/g, while for objects the size of the Earth, having the same density as that quoted, $\langle E \rangle \cong 8500$ cal/g. If all of this energy were converted to heat, the increases in the mean temperature for the two limiting cases would be ~3200°C and ~42,000°C, assuming a mean heat capacity of 0.2 cal/g/deg. Clearly, some of this energy must appear as kinetic energy of the escaping fragments; also, there is no simple way of estimating what the true mean heat capacity would be. Nevertheless, heating must have

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[1] An aeon is defined as 10^9 years, which is a convenient unit of time for discussion of the history of the solar system (*7*).

been intense indeed. One would expect the effects of this heating, e.g., severe alteration of the texture of

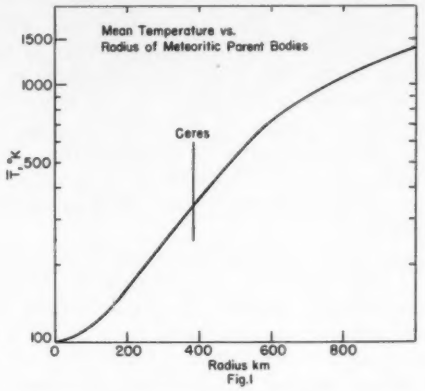


Figure 1. Volume-average temperature of meteorite parent bodies as a function of radius. The curve was calculated for bodies heated by long-lived radioactivities present in appropriate concentrations, with an initial central temperature of 1400°K and thermal conductivity of 0.007 cal cm^{-1} °K^{-1} sec^{-1}. It refers to a time two aeons after cessation of intense heating by other sources. The surface temperature was assumed to be held at 100°K by solar radiation. The radius of the largest asteroid, Ceres, is indicated for comparison.

the chondrites and of the Widmanstätten pattern of the irons (see below), to be obvious in almost all of the material which escaped the gravitational field of the combined masses.

It has recently been argued (*16*) that the chondrites must have come from parent bodies in the asteroidal belt. In addition to this theoretical argument, the only accurate, photographically determined orbit of a meteorite (the Přibram chondrite), with an aphelion of 4.1 A.U. (*17*), indicates that at least this meteorite originated in the asteroidal belt. However, it seems quite unlikely that even a single lunar-sized object could have ever existed in this region, from the small amount of material present there now [estimated as 0.03 of a lunar mass (*18*)] and the low rate at which mass can be lost from this region (*19*). Since the planetary hypothesis requires that the irons and stones should come from the same or similar parent bodies, one may cite the evidence that the iron meteorites were in fact derived from at least *four* distinct bodies[2] as indicating that it would be necessary to account for the loss of four lunar masses from the asteroidal belt, rather than one.

Perhaps the most serious difficulty encountered by the planetary hypothesis is that of constructing a thermal history of the parent body and the meteorites which fits all the evidence. The majority of chondrites display K^{40}-Ar^{40} ages of 4.0 to 4.4 aeons. It can be shown from an extrapolation of high-temperature argon-diffusion data (*2*) that these long ages imply long-term storage at mean temperatures not exceeding about 190°K (*16*). For the achondrites, only slightly higher temperatures are indicated. In a body containing "normal" amounts of uranium, thorium, and potassium, the *average* temperature due to radioactive heating is a sensitive function of size (Fig. 1), and in a body of lunar or planetary dimensions, only the topmost layers would be at the required low temperatures. The stones, comprising some 90% of observed meteorite falls, thus must be assigned to the surface layers; the irons and stony irons, representing about 10% of observed falls, are assigned to the core. One is thus forced to the paradoxical conclusion that the bulk of the parent body (subjected to pressures and temperatures too high to permit the retention of either a porous texture or the radiogenic argon content of the stones) is not represented among the known classes of meteorites.

Another aspect of the cooling problem is presented by the iron meteorites. Most of these meteorites display a characteristic structure, known as the Widmanstätten pattern (Fig. 2). It consists of broad bars of α-iron ("kamacite") and thin plates or lamellae of γ-iron ("taenite"), containing about 5–8% and 25–50% nickel, respectively. Often a finely divided mixture of the two phases ("plessite") of intermediate nickel content is also observed.

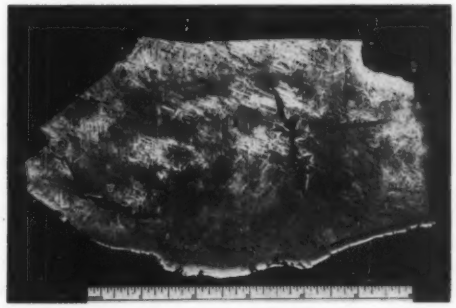


Figure 2. An etched section of the Carlton fine octahedrite (12.7% Ni), displaying the Widmanstätten pattern. Reproduced through the courtesy of Professor C. S. Smith.

The Widmanstätten pattern is usually regarded as arising from the transformation, on cooling, of the high-temperature γ-phase to α-phase or to a mixture of γ- and α-phase. For an Fe-Ni alloy containing 5.5% Ni, the γ-α transformation begins at 765°C, and is complete at 460°C. Meteorites of this nickel content (hexahedrites and nickel-poor ataxites) indeed consist only of α-phase. On the other hand, for a meteorite containing 8% Ni, transformation to the α-phase begins at 725°C, and should be complete at 280°C. Diffusion rates decrease markedly at lower temperatures, however, and at 300°C, times in excess of the age of the solar system would be required for the completion of this transformation (*22*). Hence the transformation is usually arrested at some intermediate temperature, and if the phase diagram of Figure 3 were strictly applicable, it would be possible to estimate this temperature, given the Ni-content of either the kamacite or the taenite. From this temperature, in turn, the time scale of cooling could be estimated. Unfortunately the values thus determined do not agree even within a single meteorite. In the Grant fine octahedrite, temperatures of 326° and 610°C were calculated from the kamacite and taenite, respectively

[2] Lovering, *et al.* (*20*) have shown that the gallium and germanium contents of iron meteorites do not form a continuous set, but may be divided into four well-defined clusters. The compactness of each cluster, the sharp discontinuities between them, and the decrease in gallium and germanium with increasing nickel content, in contrast to the greater solubility of these elements in nickel-rich γ-phase than in nickel-poor α-phase (*21*), all suggest that the iron meteorites originated in at least four different parent bodies.

(*23*). It appears that the binary Fe-Ni diagram is a poor approximation to the real situation, and it is of interest that Vogel (*24*) has proposed an altogether different mechanism of formation of the Widmanstätten pattern, based on the fact that the stability field of the α-phase is extended up to the liquidus in the presence of only a few tenths of one per cent of phosphorus, as is commonly found in iron meteorites. However, even in Vogel's model it is assumed that the iron meteorites cooled very slowly through the temperature range 600–400°C.

This condition presents a formidable difficulty for the planetary hypothesis. The core of the "planet" must have been molten at one time, to permit the segregation of metal and silicate, but Urey (*25*) has shown that a body of only lunar size, if melted by any process whatever, could not cool to the required low temperatures in the entire age of the solar system, even if heating by uranium, thorium, and potassium-40 is ignored. If the heating effects of these radionuclides are considered (*26*), it is found that the radius of the parent body cannot have exceeded 1000 km. It does not seem possible, therefore, to account for the detailed features of the iron meteorites by postulating their origin in planetary-sized parent bodies.[3]

Primary and Secondary Objects

The second of the principal theories of meteorite origin was conceived by H. C. Urey. He proposed that the meteorites developed in two successive generations of bodies (*6*, *7*, *22*, *25*). The first of these (the primary objects) accumulated at low temperatures, incorporating a substantial content of thermodynamically unstable compounds and possibly free radicals. Chance events, occurring from time to time, triggered exothermic chemical reactions in this material. Provided the primary object had grown to sufficient size to prevent the explosive dispersal of the reaction products (Urey has estimated that a lunar-sized object would be required), localized reduction, melting, and segregation of metal and silicate phases would result in the formation of pools of perhaps meter or tens-of-meter dimensions. The silicate pools would differentiate upon cooling into more or less pure minerals, the precursors of the achondrites, whereas the metal pools provide material for the iron meteorites. These reactions must occur near the surface, due to the difficulties in storing the reactive compounds at depth. Continued growth of the object would cause some of these segregated regions to be buried at great enough depths to permit formation of diamonds from graphite. If the temperatures in the interior of the object happened to remain near 450°C, the Widmanstätten pattern could form.

The secondary objects are thought to have been bodies of asteroidal size, or, alternatively, the surface regions of primary objects. In the secondary objects, the chondrites were agglomerated from debris resulting from collisions between primary objects, and were compacted under varying but generally low-temperature conditions. Still later in cosmic history, further collisions occurred which, in turn, broke apart these secondary objects and produced the meteoroids.

This theory has the advantage of being able to explain, or at least accommodate, most properties of the meteorites, although it often relies upon events which would seem to have low probabilities of occurrence. Some of the difficulties of the planetary hypothesis also apply to Urey's theory. If the primary objects were of lunar size or larger, the problem of mass loss from the asteroidal belt discussed above, and perhaps also the problem of breaking up such a body, must be considered. Probably the most severe disadvantage of this theory is that the range of permissible conditions in the interior of the primary object seems to be quite small. Pressures must be high enough, at some point, to synthesize diamonds, yet high pressures would lower the γ-α

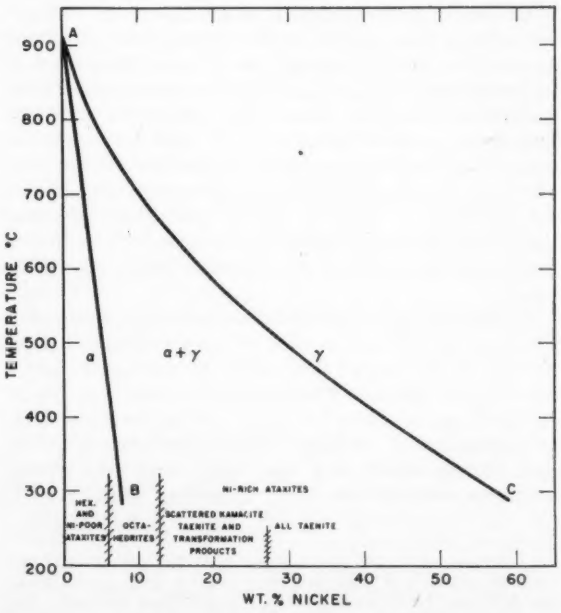


Figure 3. The Fe-Ni phase diagram, after Owen and Liu, *J. Iron and Steel Inst.*, 163, 132 (1949). Above the line AC, γ-phase (taenite) is stable, while below AB, α-phase (kamacite) is the stable form. The regions marked along the *weight* % nickel axis indicate the approximate limits of compositional variation within the different classes of iron meteorites. (Reproduced from (27) with permission of the editor.)

[3] In fairness to the proponents of the planetary hypothesis, it should be pointed out that two otherwise puzzling features of the iron meteorites may be explained by the effects of high pressures. The nickel-rich ataxites (≥16% Ni) do not display a Widmanstätten pattern, but only a rather irregular mixture of α and γ phases. The same is true of plessite (≥12% Ni) in octahedrites, which would also be expected to display a miniature Widmanstätten pattern according to Figure 3. As pointed out by Uhlig (*27*), pressure lowers the γ-α transformation temperature, so that both plessite and the nickel-rich ataxites might have been kept in the γ-form by high external pressures. Upon release of pressure during the break-up of the parent bodies, it is suggested that these alloys suddenly found themselves in the α-region, and transformed promptly to an irregular mixture of α- and γ-phase. However, this explanation of the absence of a Widmanstätten pattern in alloys of high nickel content is not unique. The phase diagrams presented by Vogel (*24*) indicate that, at phosphorus contents commonly found in iron meteorites, these alloys differ from those of lower nickel content in that there is no high-temperature α or $\alpha + \gamma$ field adjoining the liquidus in their phase diagrams. In Vogel's model, the formation of a Widmanstätten pattern would thereby be precluded. Regardless of which theory is finally agreed upon, it would seem that changes in the binary Fe-Ni phase diagram induced by the inclusion of minor constituents of iron meteorites should be carefully considered.

transformation temperature of iron-nickel alloys to the point where the diffusion rates would be small, and thus the formation times of the Widmanstätten pattern become exceedingly long. Furthermore, even if the interior of the primary object were initially as cool as 450°C, due to efficient loss of heat from near-surface chemical reactions, it would soon begin to heat up from the decay of K^{40}, U, and Th. We note also that the recent discovery of radiogenic Xe^{129} in several chondrites (*28*) implies that now-extinct radionuclides such as I^{129}, Pu^{244}, and Cm^{247}, and possibly Be^{10}, Al^{26}, and Np^{237}, would have contributed to the heating of the primary objects. It would seem that the primary objects must have been broken up very soon after their formation, both from the requirement that their interiors remain cool and from the shortness of the I^{129}–Xe^{129} decay intervals of chondrites which have been calculated [about 100 million years, according to Reynolds (*28*) and Goles and Anders (*29*)].

Two further points which bear upon the theory of primary and secondary objects may be mentioned briefly. Many polymict stone meteorites (*30*) are composed of fragments whose physical appearance and morphology are very different, but which all have the same or closely related chemical compositions. A case in point is the Cumberland Falls meteorite (Fig. 4): the fragments of black enstatite chondrite are strikingly different in appearance from the matrix of aubrite (enstatite achondrite), but chemically there is very little difference. These two classes of meteorites comprise 3.1% and 11% of the observed falls of chondrites and achondrites, respectively. If polymict stones were the products of the random collection of mixed debris by secondary objects, the probability of producing Cumberland Falls would be quite small. Similar conclusions may be reached from the study of other polymict stones. In addition, it may be noted that Cumberland Falls represents the inclusion of chondrite fragments in an achondrite matrix clearly of later origin, whereas in Urey's model the achondrites are made in the primary bodies, *before* the synthesis of chondrites.

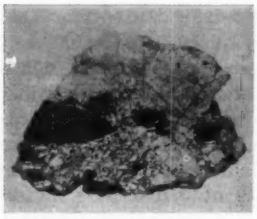


Figure 4. Cumberland Falls meteorite, a polymict, brecciated stone. Fragments of black enstatite chondrite are embedded in a matrix of white aubrite (enstatite achondrite). Evidently, the chondrite cannot be of later origin than the achondrite. (Reproduced through the courtesy of Dr. E. P. Henderson of the U. S. National Museum.)

Asteroidal-Sized Parent Bodies

Numerous authors have suggested, during the last century, that the asteroids or "minor planets" were the immediate parents of the meteorites. All of the larger asteroids are spherical (*31*), indicating that they are not fragments of a larger planet, but had formed as independent bodies. The smaller asteroids, on the other hand, often display irregular light curves (*31*), which are best interpreted as due to their origin as irregularly-shaped fragments of larger objects. The collision lifetimes of typical asteroids have been estimated to be on the order of one aeon (*19*), and the annual production of dust and rubble from collisions within the asteroidal belt has been calculated to be $\sim 10^{12}$ kg, more than ample to account for the apparent frequency of meteorite infall on the earth's surface (*32*). Thus, it seems quite reasonable that the material in the asteroidal belt is the immediate source for most, if not all, of the meteorites.

It is tempting to speculate that the asteroids are not merely the last residence of the meteorites, but represent the parent bodies in which these objects acquired their structures. However, a detailed analysis of this suggestion reveals three important difficulties. The chief of these, as pointed out by Urey (*6*, *7*, *33*), is that of the nature of an energy source intense enough to provide heat for melting portions of the meteoritic parent bodies. Temperatures in excess of 1800°K, the melting point of iron, are required for separations of iron and silicate phases: forsterite inclusions in the Tucson meteorite imply temperatures of ~2100°K; and both the flow structures of the orvinites and the preterrestrial magnetic moments of some chondrites (*34*) indicate that temperatures in the interiors of the parent bodies of these meteorites were of the same magnitude. The surface-to-volume ratio of asteroidal-sized bodies is so large that heating by long-lived K, Th, and U radioactivities could not have provided central temperatures above ~1000°K, even in the largest of the asteroids (Ceres, radius = 385 km). Also, the surface gravity of such a body is too low to permit the effective operation of the Urey-Donn chemical heating mechanism.

The two remaining difficulties are connected with the small gravitational potential of asteroidal-sized objects. It has been suggested (*22*) that the presence of diamonds in meteorites is indicative of pressures on the order of 3×10^4 atm, more than a factor of ten greater than the gravitational pressures of asteroidal interiors. Finally, it has often been claimed that gravitational phase separations would be inhibited in small bodies.

The last objection, that gravitational separations of iron and silicates and of one silicate phase from another would be inefficient in small bodies, was critically examined by R. A. Fish and the present authors (*35*).

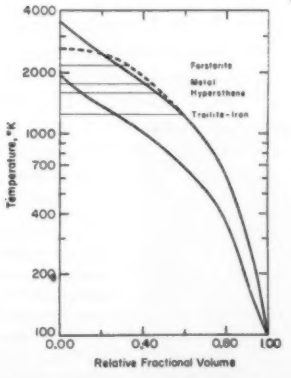


Figure 5. Interior temperatures of radioactively heated planetesimals versus fractional volume. The lower curve is a typical curve for planetesimals in which central melting has just begun. The solid portion of the upper curve is representative of planetesimals with superheated cores; no allowance has been made for convective heat transport or other perturbations. The dashed portion of the upper curve is an estimate of the temperatures in the case that convection in the molten central regions and conduction in the inner metal core are both effective. The horizontal lines indicate melting points of various meteoritic phases. [Reproduced from (35). Copyright (1960) by the University of Chicago.]

It was demonstrated that the efficiency of gravitational separations is not a function of the total radius of the body, but only of the distance of the differentiating melt from the center of the body. It may be shown that the observed meteorite phase separations could easily be attained in kilometer-sized objects, if an appropriate heat source were present.

Fish *et al.* (*35*) proposed that the remaining objections to the hypothesis of the origin of meteorites in small bodies could be tentatively set aside by means of two assumptions: That a transient, internal heat source, such as extinct radioactivity (*33*, *36*), had been present, and that meteoritic diamonds could have been produced without high gravitational pressures.[4] Granted these premises, the thermal history of asteroids heated by such a source was investigated in order to discern the spontaneous processes that might occur in these bodies. It then remained to be shown that these processes sufficed to account for the principal features of meteorites.

The temperature distribution in an internally heated asteroid is given in Figure 5. Reduction of Fe^{++} to the metal will set in at temperatures of 600°K, and once melting begins at the center, the body will consist of an inner core of metal, an outer core of molten silicates, a sintered mantle, and an unconsolidated surface layer. This stratification could account for the composition and texture of the known classes of meteorites: the materials of the highly differentiated irons, pallasites, and achondrites would come from the core; the compact, well-sintered mesosiderites and crystalline chondrites, from the deeper regions of the sintered mantle; the more friable types of chondrites, from the intermediate and upper layers; and the carbonaceous chondrites, from the consolidated surface regions.

An interesting consequence of this model is that it provides for the operation of a quasi-volcanic process. On heating, the following reactions could take place in the core:

$$FeS + SiO_2 + 2C \rightarrow Fe + SiS + 2CO \quad (2)$$

$$2FeS \leftrightharpoons 2Fe + S_2 \quad (3)$$

$$Fe + SiO_2 \leftrightharpoons SiO + FeO \quad (4)$$

The first of these is used commercially in the desulfurization of steel, and gives a silicon monosulfide partial pressure of one atmosphere at temperatures as low as ~1800°K (R. M. Fowler, private communication). The second and third reactions require higher temperatures, of about 2500 and 3000°K, in order to develop comparable pressures. With the amounts of carbon and iron sulfide normally present in iron meteorites, reaction (2) should take place continuously at the boundary between the metal and silicate layers in the molten core. If the mantle is porous enough to be permeable to gases, the gaseous products will flow outward; SiS will undergo chemical reactions or condense in the cooler regions, whereas CO will largely escape even if it is partly oxidized to CO_2. On the other hand, if the deeper portions of the mantle have been sintered sufficiently to make them gas-tight, the SiS and CO will accumulate in the core until their combined pressure exceeds the sum of the tensile strength and the gravitational pressure of the overlying layers. At that time, an explosive, quasi-volcanic eruption may occur.

An eruption of this kind could carry molten material to the surface, where it would be dispersed by turbulence into rapidly solidifying droplets. It is interesting to note that many students of meteorites have previously concluded that some kind of volcanism would best account for many structural features of the meteorites. In particular, the chondrules are almost universally believed to have originated by dispersal and rapid cooling of a melt (*15*, *41*, *42*).[5] Volcanism of the terrestrial type, based on water vapor, is not likely to have been of importance in asteroids, since low-boiling substances of low molecular weight would be lost from the interior of the object at an early stage. The model proposed obviates this difficulty by allowing for the generation of gases at high temperatures from nonvolatile substances.

A desirable feature of this process is its cyclic nature. After each eruption, the fissures in the mantle will be sealed by sintering or by intrusion and freezing of a melt, so that, as long as the heat source persists, pressures can again build up in the core. The amazingly complex, brecciated structures of many stone meteorites, exhibiting up to five generations of fragments within fragments (*30*), seem to require repeated cycles of heating, chondrule formation, fragmentation, reaccumulation, and mixing.

The "proof" of any such hypothetical model must, of necessity, rely largely on circumstantial evidence. It is therefore essential that the physical conditions and processes proposed in the model be consistent with all known properties of meteorites. Unfortunately, few meteoritic properties exist for which there is a single, unique explanation; in some cases, however, one model may offer a simpler and less contrived explanation than another. Two such cases, for which the model under discussion seems to provide a relatively straightforward rationale, are the capillary veins in stone meteorites, and the abundances of some heavy elements in chondrites. By discussing these properties in detail, we hope to illustrate the type of reasoning that is generally used in this field. These properties are by no means the only ones of significance, but they were established only recently, and have therefore not been as fully discussed in the literature as some others.

Many stone meteorites are permeated by a network of veins, ranging from a centimeter to about 0.01 mm

[4] Recently, both of these assumptions have received support from experimental evidence. The suggestion that extinct radionuclides were present during the synthesis of meteorites was confirmed by the discovery of radiogenic Xe^{129} in meteorites by Reynolds (*28*, *37*). Theories for the incorporation or production of these radionuclides in the solar nebula have recently been proposed (*38*, *39*). However, it still remains to be demonstrated experimentally that any of these radionuclides, e.g., Al^{26}, were present in amounts sufficient to act as major heat sources. With respect to meteoritic diamonds, Lipschutz and Anders (*40*) have presented a detailed argument, based on metallographic evidence and thermodynamic calculations, that the diamonds in the Canyon Diablo meteorite were formed on impact with the earth. Very little is known concerning the mode of occurrence of diamonds in other meteorites, but it has been suggested also that they were formed by shock waves, either during impact with the earth or during the disruption of the parent body.

[5] This explanation is not unique, however. Urey and Craig (*5*) have suggested that chondrules originated in collisions, Ringwood (*12*) believes that they crystallized *in situ*, and Levin (*43*) proposed that they represent primary condensates in the solar nebula.

in thickness (Fig. 6). A most important clue to their origin was provided by the Viennese geologist Berwerth in 1903 (*44*). In his study of the Peramiho eucrite, he states:

> The compositional changes within a single vein are highly instructive and of importance in establishing the nature of veins. A fine, black vein that can be traced over considerable distances suddenly terminates at a feldspar grain, and then resumes at the other side of the crystal. This break is only apparent, however, since the vein is not actually interrupted by the feldspar, its continuity being restored by [a band of] white anorthite (feldspar) glass. All of the above phenomena indicate that the veins were produced within the stone by the effect of heat.

To a chemist, this observation is extremely suggestive. The narrow width and glassy character of the

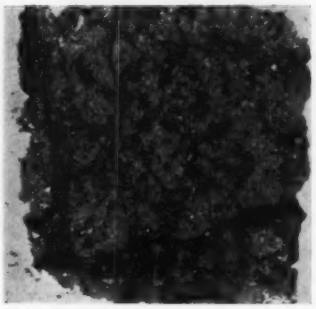


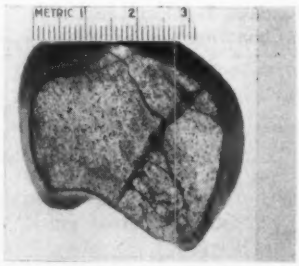
Figure 6. Veins in the Walters stone meteorite. Note the wide range in vein thickness and also the complex, branching network of capillary veins. (Reproduced through the courtesy of Dr. S. K. Roy, of the Chicago Natural History Museum.)

vein in the anorthite crystal seems to imply very brief and localized melting, followed by rapid cooling. But why is the vein black in the pyroxene [$(Mg^{++}, Fe^{++})_2Si_2O_6$] matrix and colorless in the anorthite [$CaAl_2Si_2O_8$] crystal? Some material must have been introduced that gave a black reaction product with the pyroxene but not with the anorthite. Could it be that the iron content of the pyroxene was responsible for the dark color?

Compounds to be considered as the source of this coloration are: finely divided metallic iron, magnetite, and troilite (FeS). For the latter compound, a simple metallographic test is available, the so-called "sulfur print." The polished specimen is placed on a piece of wet photographic paper that has been soaked in dilute sulfuric acid. Any acid-soluble sulfide will react with the silver bromide in the emulsion, e.g.:

$$FeS + 2AgBr \rightarrow Ag_2S + Fe^{++} + 2Br^- \qquad (5)$$

producing a sharp contact print which reveals the distribution of sulfides in the specimen.



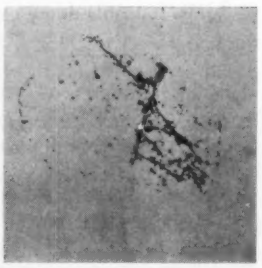


Figure 7 (left). Section of Harrisonville chondrite with prominent vein system.
Figure 8 (right). Sulfur print (see text) of Harrisonville section of Figure 7. Areas rich in troilite show up dark in print; note that veins are enriched in troilite. [Reproduced from (35). Copyright (1960) by the University of Chicago.]

The first veined meteorite that was tested in this manner was the Harrisonville, Missouri, chondrite (Fig. 7). We found that the X-shaped vein pattern was indeed accurately reproduced on the sulfur print (Fig. 8), indicating a substantial local enrichment of sulfide in the veins. We have since tested numerous other veined meteorites, and found the same enrichment in sulfide in every instance.

Under the microscope, a number of additional features of veins become apparent. The veins can be plainly traced by following the stream of finely divided troilite particles (Fig. 9), which often coalesce to round droplets (Fig. 10). These are suspended in glassy-appearing silicates, implying that temperatures above

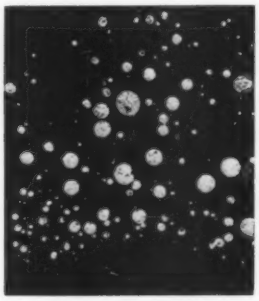


Figure 9. (left). Enlargement of vein and surrounding regions of Harrisonville section. Note troilite droplets scattered along vein, especially near metal fragments.
Figure 10 (right). Further enlargement of troilite droplets in vein. The apparently spherical shape indicates that their silicate matrix was once molten, as does the glassy character of the silicate. Shearing flow must have been largely absent during solidification.

~1800°K were reached, yet only a few millimeters away are found single-crystal kamacite grains with Neumann lines, features that would not survive heating above ~1000°K for even a few seconds. An even more striking case is presented by the split taenite grain in Figure 11. Originally, this particle must have been completely surrounded by a dark "diffusion border" of very fine-grained α-γ mixture, which is believed to have formed on slow cooling of the γ-alloy in the $\alpha + \gamma$ field. On reheating, the border disappears, due to conversion of α-phase to γ (see Fig. 3), and if the reheating is followed by rapid cooling, the particle will

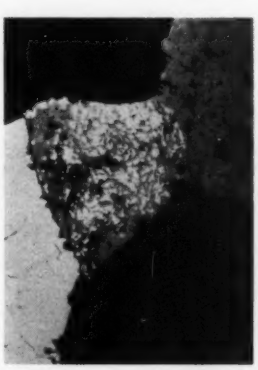
Figure 11. Split taenite (γ-phase) grain traversed by a vein. The discontinuous diffusion border around the particle indicates that it was once kept at temperatures of some 800–1000°K, where a continuous border could develop, and that it was subsequently fractured and was then exposed briefly to high temperatures. After that, it remained at temperatures too low to permit the redevelopment of a continuous diffusion border. The granular structure is identical with that obtained upon reheating of plessite to about 900°K. The persistence of some needle-like plessite in the particle on the left, which lies farther from the vein, implies that the heat was applied for so short a time that even a thin band of silicate could act as an effective thermal barrier. The abrupt appearance of troilite droplets in the vein, beginning in the center of the photograph, is apparently due to reaction of hot sulfur-containing vapors with the metal particle. [Reproduced from (35). Copyright (1960) by the University of Chicago.]

become polycrystalline and granular in appearance. Precisely this effect may be observed. The border has survived in the particle on the left, on the side farthest from the vein, and at one edge of the fragment on the right, but has disappeared everywhere else. Similarly, the original needle-like plessite structure has survived in the region farthest from the vein but has disappeared elsewhere, giving way to the granular structure characteristic of reheated metal. Evidently the reheating in this instance must have been of exceedingly short duration, so that the heating effects were localized to the volume within hundredths of a millimeter from the vein. One should note that other meteorites, with larger veins, exist in which heating effects are traceable for as far as several centimeters from the veins.

Meteoritic veins could not have been produced by frictional heating. Aside from the fact that this mechanism would not account for the observed enrichment in sulfide, the heat available is clearly insufficient. The frictional force is equal to the force pressing the surfaces together, times the frictional coefficient. A generous upper limit to the pressure may be obtained by setting it equal to the compressive strength of the hardest chondrites, ~3500 kg/cm² (*45*), and the frictional coefficient may be taken as 0.7 (a typical value for stone and masonry). For these values, we find that the frictional force is 2.4×10^9 dynes/cm.² The lateral displacement of the vein walls occasionally is as great as 0.1 cm (for the large veins), as measured from the relative displacement of the portions of a split metal particle or chondrule. For such a displacement, the work done against the frictional force is 2.4×10^8 ergs/cm², or ~5 calories/cm². In a vein of 0.1 cm thickness, the temperature will rise by merely 75°C, if the specific heat of the walls is taken to be 0.2 cal g^{-1} deg^{-1}, and their density, 3.3 g/cm³. Obviously, this is far from sufficient to account for the observed heating effects. In the finest capillary veins, with diameters as small as 10^{-3} cm, fusion would indeed occur at pressures of 3500 kg/cm², if the displacement were as large as 0.1 cm. But in none of the capillary veins examined have we ever found displacements even as large as 2×10^{-3} cm, corresponding to a temperature rise of ~150°C. Moreover, the capillary veins often form closed loops, or are right-angle branches of larger veins, so that extensive lateral displacements are impossible for geometric reasons.

It can also be shown that the veins cannot have been caused by injection of a melt. The flow of any such melt must have been extremely rapid, to allow complete penetration of capillary cracks before enough heat had been conducted to the walls of the cracks to cause the melt to freeze. Flow rate depends on the head pressure, the viscosity of the melt, and the dimensions of the vein system. There is no reasonable choice of the pressure and viscosity such that a melt can be injected fast enough to meet the above condition, but for a gas or superheated vapor, with viscosities some 10^{-6} to 10^{-8} that of a liquid, the situation is far more favorable. By a process of elimination, one is thus led to the conclusion that the veins were produced by the momentary passage of hot, sulfur-containing gases or vapors through cracks in the mantle of the parent body. It is of interest to recall that the explosive release of hot, sulfur-rich vapors has been postulated on independent grounds, as part of the volcanic process in the asteroidal-sized parent bodies described by Fish, *et al.* (*35*).

Recently, Hentschel (*46*) concluded from a study of the Breitscheid chondrite that part of the troilite in this meteorite was of "secondary" origin, having been formed by partial replacement of metal by troilite. In contrast to primary troilite, secondary troilite contains small irregular grains of metal, possibly representing the nickel-rich residue from the sulfuration of the metal (Fig. 12). Similar grains are visible in Figure 13, from the paper by Fish, *et al.* (*35*).



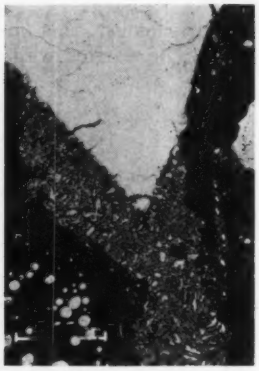


Figure 12 (left). Primary and secondary troilite in Harrisonville. The granular primary troilite (gray area in upper right) adjoins secondary troilite (mottled gray-white area in center) which was apparently derived by sulfuration of the contiguous kamacite (white area, lower left). Note that the kamacite is polycrystalline, implying reheating late in its history.
Figure 13 (right). Kamacite particle adjoining vein. Note concentration of troilite droplets and metal fragments at lower end of particle. The appearance of this area suggests corrosive attack of the metal particle by hot sulfur vapors. [Reproduced from (35). Copyright (1960) by the University of Chicago.]

Another line of evidence in support of this hypothesis comes from studies of the distribution of certain trace elements in chondrites. Reed, *et al.* (*47*) and Ehmann and Huizenga (*48*) showed that the abundances of thallium, lead, and bismuth in bronzite and hypersthene chondrites are far lower than would be expected, either from theories of nucleosynthesis or from the concept of a smoothly varying abundance curve (Table 1). It might be argued that these elements, being the heaviest of the stable ones, perhaps should not follow the predicted trends. For instance, the abundances of their neutron-rich progenitors in the *r*-process of nucleosynthesis might be abnormally low, due to the

Table 1. Abundances of Indium, Thallium, Lead, and Bismuth in Chondrites

	Hypersthene and bronzite chondrites		Enstatite and carbonaceous chondrites (range)	Suess-Urey abundances (*4*)
	Range	Median or av.		
In	.....	<1 ppb (*49*)	.....	80.0 ppb
Tl	0.03–3.8 ppb (*47*)	~0.4 ppb (*47*) 1.3 ppb (*48*)	70–140 ppb	140 ppb
Pb	0.04–0.6 ppm (*47*)	~0.15 ppm (*47*)	1–5 ppm (*47*)	0.62 ppm
Bi	0.4–8 ppb (*47*)	~3 ppb (*47*) 2.2 ppb (*48*)	80–180 ppb (*47*)	190 ppb

effect of the closed nuclear shell at 126 neutrons. However, Schindewolf and Wahlgren (*49*) found the abundance of indium, an element in the middle of the periodic table, to be nearly one hundred times lower than expected. There is no nuclear property of indium that would account for its rarity, and one is therefore led to believe that a chemical fractionation process must have been responsible. Evidence in support of this view was recently furnished by Reed, *et al.* (*47*), who found that the "deficient" elements thallium, lead, and bismuth are present in nearly their expected abundances in members of two rare subclasses of chondrites, the carbonaceous and enstatite chondrites.

Urey (*9, 11, 50*) has discussed chemical fractionations in meteorites by volatilization, and pointed out that mercury, due to the high volatility of its compounds, would be the first of the so-called "non-volatile" elements to be lost upon heating, followed by zinc and cadmium.[6] It is therefore puzzling, at first sight, that the more volatile elements (including mercury) are not greatly underabundant in bronzite and hypersthene chondrites, whereas some elements of lower volatility are very strongly depleted (Fig. 14). Evidently, the fractionation was not caused by simple volatilization.

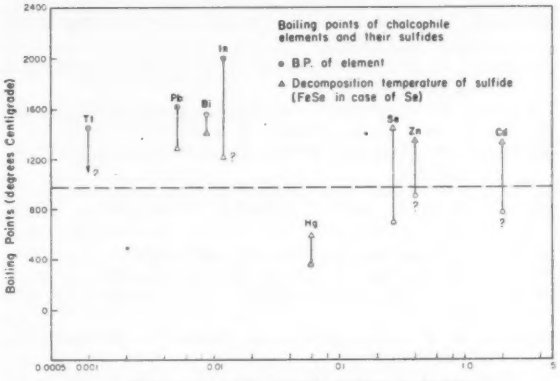


Figure 14. Boiling points of chalcophile elements and their sulfides versus depletion factors in chondrites. The relatively non-volatile elements Tl, Pb, Bi, and In, whose boiling points are higher than the decomposition temperatures of their sulfides, are strongly depleted, whereas the volatile elements Hg, Se, Zn, and Cd are much less affected, if at all.

The "calculated" abundances of In, Se, Zn, and Cd were taken from Suess and Urey (*4*) and those of Tl, Pb, Bi, and Hg, from Cameron (*54*). The observed abundances are from various recent studies (*47, 48, 49, 55*). The abundances of Zn and Cd, and the decomposition temperatures of thallium and indium sulfides are uncertain.

A clue to the resolution of this paradox is actually provided in Figure 14—for the strongly depleted elements, the sulfide has a higher boiling point or decomposition temperature than the element itself, whereas the reverse is true for the less strongly or non-depleted elements. The observed fractionation could be explained in the following way: In an internally heated planetesimal of chondritic composition, the first major phase to melt will be the troilite-iron eutectic (mp 1261°K). Owing to its high density, the eutectic melt will flow toward the core, dissolving elements that form stable sulfides at or above this temperature. Reference to Figure 14 indicates that all the depleted elements, with the possible exception of mercury and thallium, will be extracted. (Actually, as a consequence of Henry's Law, the decomposition temperatures of the diluted sulfides will be higher than the values for the pure compounds indicated in Figure 14, so that even mercury and thallium are likely to be extracted.)

In its flow toward the core, the Fe-FeS eutectic will percolate through regions of progressively higher temperatures. Eventually the temperature will become high enough to decompose the dissolved trace metal sulfides, most of which have similar decomposition temperatures. At this stage, the great difference in boiling points of the two groups of elements will become important. The partial pressure of sulfur vapor above the melt is fixed by the FeS decomposition equilibrium, and is held to a low value due to the excess of free iron in the eutectic. The partial pressures of the metal vapors, on the other hand, are given by the respective Henry's law constants, which for very dilute solutions should be roughly proportional to the vapor pressures of the pure metals. Manifestly, these pressures will be enormously greater for the more volatile of these elements, e.g., mercury, cadmium, and zinc. If the vapor pressures exceed the confining pressures, these elements will be returned to the chondritic mantle. In view of the thermal gradient which exists in the mantle, these elements may be expected to condense in the inverse order of their boiling points, although they may be partially retained in warmer regions by reaction with iron sulfide grains. Mercury in particular should condense largely in regions close to the surface, and it is interesting to note that in the single carbonaceous chondrite studied, Reed, *et al.* (*47*) have indeed found the remarkably high mercury content of 10^4 ppb. This is in agreement with the assignment of carbonaceous chondrites to the cold surface regions of the parent planetesimals.

If this picture is correct, then only those meteorites that consist largely of materials once subjected to temperatures above 1261°K should show this fractionation. The carbonaceous chondrites appear to have spent most of their history at temperatures below 500°K, and it may be that they have thus retained the primordial abundances of these trace elements, with the exception of mercury. The low mercury content and other properties of the enstatite chondrites are definite evidence of a high-temperature history, but the constancy of their Ni/Si ratios (*5*) and the incomplete segregation of metal and silicate (*52, 53*) suggest that the maximum temperatures to which these meteorites were subjected were not high enough to cause extensive fractionation by the mechanism discussed above.

This model is still inadequate in some respects. The troilite content of chondrites is remarkably constant at about 5% (*5*). This troilite cannot simply be an aliquot of the original troilite, since an initial content of 10^2 to 10^3 times the observed amount would be required to account for the large depletion factors of thallium and bismuth. A mechanism is apparently needed for the return of sulfide, fractionated with respect to the trace elements, to the mantle. Such a mechanism is provided, in principle, by reactions (2)

[6] Arsenic and antimony were also included in Urey's original list, but since these elements, just as their congener phosphorus, form relatively non-volatile alloys or compounds with iron and nickel, their volatilities are much lower than previously estimated. In fact, arsenic is known to occur in the metal phase of chondrites in approximately the expected abundance (*51*).

and (3) given above. If either sulfur vapor or silicon monosulfide were to undergo chemical reactions in the mantle, e.g.:

$$S_2 + 2Fe \rightarrow 2FeS \qquad (6)$$

or

$$SiS + 2Fe_2SiO_4 \rightarrow 2FeSiO_3 + FeS + Fe \qquad (7)$$

the "solvent" FeS would be regenerated, and a cyclic process resembling a Soxhlet extraction could take place. However, much experimental work, particularly on the high-temperature behavior of an Fe-FeS melt containing these trace elements and in contact with other meteoritic phases, will be required to establish the validity of this model.

While the hypothesis of meteorite origin in a single generation of asteroidal-sized bodies appears to be capable of providing valuable insights, it has not yet been directly proved whether a heat source of the required type, e.g., Al^{26}, was present during the early history of the solar system. Furthermore, this hypothesis does not seem to provide an explanation for the variations in mean density between the earth and the moon, and among the inner planets (*8*, *25*). These variations do not have a direct bearing on the history of the meteorites, but any theory of the origin of meteorites should ultimately be tested for consistency with our knowledge of the solar system as a whole.

Conclusions

We have presented brief outlines of the three principal theories of meteorite origin. Although the problems involved have received much careful scrutiny from workers in various fields, none of these theories can at present be considered to be definitive. This situation is due, in general, to two factors: the material under study is very complex, and the phenomena, because of obvious limitations, cannot be reproduced in the laboratory. Nevertheless, it is possible to treat this subject in a rigorous manner, being guided by the principles that *all* the pertinent data should be considered and that the models proposed should be self-consistent and involve, as far as possible, only processes of reasonable probability. With this in mind, the major aim of our discussion has been to stimulate interest in devising crucial experimental tests for the theories outlined, largely by indicating the way in which some experiments have affected theory. It is certain that our concepts of the origin of meteorites will undergo many changes in the next few years. One would hope that our understanding of the meteorites will be increased and that, eventually, it will be possible to construct a detailed history of these most complex and fascinating objects.

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The Chemistry of Creation of the Heavy Elements

Chemists have been interested from time immemorial in the chemical composition of the world around us. With the growth of knowledge about nuclear transmutation and about nuclear reactions in stars, it is now possible to outline the main processes by which our elements were made from the simplest one, hydrogen. The present treatment will be restricted to the main process responsible for the heavy elements, of mass number A greater than 70. The process is that of very rapid capture of fast neutrons in a supernova explosion, whereby atoms in the iron region (where nuclear binding is tightest) suddenly are exposed to a tremendous flux of fast neutrons of short duration, and hundreds of neutrons may be captured. Subsequent radioactive decay of these products gives us the elements we now know.

Nuclear Shell Structure

The nucleus is composed of Z protons and N neutrons in very tight combination of mass number $A = Z + N$. The typical binding energy, B^p or B^n for protons or neutrons, is 6–8 Mev for the species stable to β^- decay of electron capture. There is a well-defined pattern (*1*) of Z_{stab} and N_{stab} for the 300-odd β-stable species known in the region $1 \leqslant A \leqslant 260$. Nuclides of given A with N values lower than N_{stab} in this pattern change protons into neutrons by electron capture; those with N values higher change neutrons into protons by β decay. All nuclides with $Z > 83$ (and a few others) undergo α decay, but some of these in the region near $Z = 92$ have rather long half-lives for α decay. Three of these, Th^{232} (1.4×10^{10} yr), U^{235} (7.13×10^8 yr), and U^{238} (4.51×10^9 yr) have half-lives comparable with the age of the elements and, like several β unstable low-Z species (e.g., 1.3×10^9-yr K^{40}), they are still found in measurable amounts.

It is taken as an axiom that all stable nuclei are found in nature, and that all other combinations of Z and N will be unstable. Species of half-period longer than about 5×10^8 yrs are also found in nature, having persisted since the time of nucleosynthesis.

Quantum mechanical considerations control the interactions of the Z protons and N neutrons of the nuclide Z^A. Coulombic repulsion tends to keep the protons far apart, favoring high angular momentum quantum-number l, although the intrinsic nuclear proton-proton attraction is as high as the neutron-neutron attraction. The high neutron-proton interaction (not affected by the Pauli Exclusion Principle) tends to keep the neutrons also in states of high l. As pointed out by Mayer (*2*) and Haxel, *et al.* (*3*), the final key to nucleon shell structure is the recognition of high spin-orbit coupling. For a state of high l, the quantum configuration with nucleon spin $^1/_2$ parallel to l, of total angular momentum $j = l + {}^1/_2$, is of lower energy than that of $j = l - {}^1/_2$ by about 2 Mev for large l values.

Shell-closure in the nucleus, as in the electronic structure of the atom, is a consequence of a gap in energy levels. Shell-closure in nuclei occurs when either Z or N attains the numbers,[1] 2, 8, 20, 28, 50, 82, or 126. These numbers correspond to the population permitted by the Pauli Exclusion Principle in the filling of nucleon orbitals up to the respective points where bars occur in the following set[2]

$$1s \,/\, 1p,\ 1d,\ 2s \,/\, 1f_{7/2} \,/\, 1f_{5/2},\ 2p,\ 1g_{9/2} \,/\, 1g_{7/2},\ 2d,\ 3s,\ 1h_{11/2} \,/\, 1h_{9/2},\ 2f,\ 3p,\ 1i_{13/2} \,/\, 1i_{11/2} \ldots$$

In the orbital of highest energy, neutrons remain paired as far as possible, as do protons. This is a contrast with electronic structure (Hund's Rule), where electrons in the highest orbital keep the maximum number unpaired that is permitted by the Pauli Exclusion Principle. The pairing energy of two neutrons or two protons is 1.5–2.5 Mev, depending on the shell region. It explains the great preponderance of β-stable even-A nuclides of even-Z, and the total absence of β-stable even-A isotopes of odd-Z elements beyond $_7N^{14}$. All even-Z even-A species have zero spin; odd-A nuclides generally show the spin properties of the odd proton or odd neutron [the Independent Particle Model (*4*)].

Nuclear shell structure corresponds to a drop of about 2 Mev in a binding energy of about 8 Mev for common nuclei, which is much less pronounced[3] than electronic

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[1] The shell numbers quoted here were found empirically, and were called "magic numbers" before they were explained on quantum mechanical principles.

[2] The orbitals are designated s, p, d, f, g, h, i, j for l values of 0, 1, 2. . . 7. The subscripts refer to the j values, $l \pm {}^1/_2$. The successive recurrence of l values is designated by the numbers 1, 2, 3, . . .to mark the recurrence of a given l value, as in numbering levels for a harmonic oscillator. The first time $l = 3$ occurs, for instance, it is called $1f$, not $4f$ as in electronic nomenclature.

[3] The drop in electron binding energy (ionization potential) is from typically 15 ev for a noble gas to 5 ev for an alkali metal. The order of electronic shell structure is set by minimum angular momentum, in order to bring each electron close to the nucleus. Shell structure is set by the gap in energy following filling of the $1s$ level, and after that the successive p levels $2p$, $3p$, $4p$, etc. The shell numbers are: 2, 10, 18, 36, 54, 86, and 118; these are the atomic numbers of the noble gases.

shell structure. It does however make a noticeable effect in nuclear properties, especially dynamic ones. The so-called "valley of stability" in the nuclear energy surface is narrowed in the region of shell number for Z or N, leading to a small favoring of shell numbers in the pattern of stability. The fall in binding energies on crossing a shell number is quite apparent.

The most dramatic effect, however, is on reaction probability. Nuclides with closed shells of neutrons or protons show marked fall in cross section σ for adding fast neutrons (*5*) compared to other nuclides in the same general A region. This fall is approximately 50 fold, apparently observed only for those nuclides available for study with closed proton or neutron shells. In other features, the cross sections for fast neutrons ($\sim$1 Mev) increase exponentially with A until A is about 100, after which they remain constant at about 0.1 barn (10^{-24} cm^2/atom) for the rest of the region available for study.

Astronuclear Reactions

Most of the matter in the universe is hydrogen (*6*), and astrophysical models of stellar reactions (*7*, *8*) consider stellar development beginning with $_1H^1$ as the raw material. When sufficient hydrogen is collected gravitationally ($\sim 10^{33}$ g), temperatures rise from the release of gravitational potential energy, heating the core to a temperature of about 10^7°K. Nuclear energy then is released by the thermonuclear reactions:

$$_1H^1 + {}_1H^1 = {}_1H^2 + \beta^+ + \nu \qquad (1)$$

$$_1H^2 + {}_1H^1 = {}_2He^3 + \gamma \qquad (2)$$

$$_2He^3 + {}_2He^3 = {}_2He^4 + 2\,{}_1H^1 \qquad (3)$$

at a rate set by that of equation (1) and with a net energy release of 26.7 Mev for the over-all reaction:

$$4\,{}_1H^1 = {}_2He^4 + 2\beta^+ + 2\nu \qquad (4)$$

If some $_6C^{12}$ is present in the stellar core, another reaction, the Bethe cycle, gives a higher conversion rate at slightly higher temperatures. This involves the successive reactions $C^{12}(p,\gamma)N^{13}(\beta^+)C^{13}$; $C^{13}(p,\gamma)N^{14}$; $N^{14}(p,\gamma)O^{15}(\beta^+)N^{15}$; and $N^{15}(p,\alpha)C^{12}$, with the same over-all equation as (4).

If sufficient great mass is available in the star, gravitational energy will drive the core to still higher temperatures when all of the $_1H^1$ is burned out. At temperatures around 2×10^8°K and densities of about 2×10^4 g/cm^3, helium nuclei condense to carbon nuclei:

$$3\,{}_2He^4 = {}_6C^{12} \qquad (5)$$

releasing 7.2 Mev. At still higher temperatures and pressures, other heavy nuclei are formed, until for the heaviest stars, the cores can go to thermonuclear equilibrium with large amounts of $_{26}Fe^{56}$ present. No further nuclear potential energy is available to make heavier elements, as the nuclear stability falls with A for heavier nuclei.

Some neutrons can be made in the previous types of syntheses if protons are brought from outer shells into a hot core with helium and heavier elements. The protons will be changed into nuclei like C^{13}, Ne^{21}, Mg^{25}, and the (α, n) reaction on these produces neutrons.

A star that is heavy enough can condense nuclei until most of its core material is in nuclei in the region of iron. It will be at very high density ($\sim 10^6$ g/cm^3) and temperature ($\sim 5 \times 10^9$°K). A sharp change in the matter in statistical equilibrium sets in under further gravitational action, leading to breakdown:

$$_{26}Fe^{56} = 13\,{}_2He^4 + 4\,{}_0n^1 \qquad (6)$$

This reaction is endothermic by 124.5 Mev. It leads to a spectacular collapse of the core with a time scale of only minutes. The outer layers also collapse with great heat output, leading to a supernova explosion. Part of the mass is blown off into space at high velocity. Supernova explosions are thought to occur about once per 300 years per galaxy. Our Crab Nebula is the product of such an explosion in 1054 A. D.

Neutron Capture and Isotopic Abundances

The abundances of the different naturally occurring isotopes of elements are known with high precision. No significant difference is found for samples from different terrestrial sources, or between terrestrial samples and meteoritic samples. There is a general consistency in the pattern for all heavy elements, similar to that shown in Figure 1. This is a plot of the isotope abundances of stable species with Z values between 44 and 49 and N values between 56 and 68. Stable species are outlined in black with mass number A and per cent isotope abundance in the square. For interest, half-lives are shown for some of the unstable nuclides. Neutron capture corresponds to movement one space to the right; β decay corresponds to movement diagonally up one and one to the left.

This pattern finds simple explanation if neutron capture, in a steady state, with β decay of unstable species, was a common step. If the neutrons come on

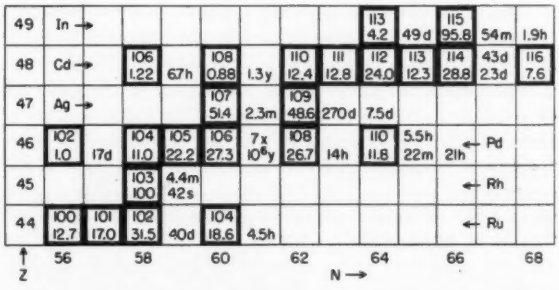

Figure 1. Typical pattern of isotope abundances above mass number 80. Stable nuclides are shown as black-edged squares with mass number and isotope abundance in per cent. Half-lives are shown for relevant unstable species: *m* = minutes, *h* = hours, *d* = days, *y* = years. *Ordinate:* atomic number *Z*; *abscissa:* neutron number *N*.

a slow time scale, with repetition in times large compared to 40 days, but small compared to 7×10^6 yr (Pd^{107}), the reaction path would include Ru^{100}–Ru^{102}, Rh^{103}, Pd^{104}–Pd^{108}, Ag^{109}, Cd^{110}–Cd^{114}, In^{115}. The subsequent β decay of 7×10^6-yr Pd^{107} would give an equal amount of stable Ag^{107}. All stable nuclei to the left of this line (except Ag^{107}) occur in rather low abundance, and are presumably made by photonuclear processes and proton capture. Fowler (*7*, *9*) calls neutron capture on a slow time scale the *s* process, and the rare mode of production of the light even-Z isotopes the *p* process.

The heaviest stable isotopes of even-Z elements such as Ru^{104}, Pd^{110}, and Cd^{116}, could be made by neutron

capture only if the repetition time is short compared to the half-lives of the intermediate β-unstable species Ru^{103}, Pd^{109}, and Cd^{115}. Such a rapid capture process is called the *r* process (*7*, *9*). The abundances of the stable species formed by it indicate that it was a prominent process, but give no obvious measure of the timing of successive neutron capture on a given nucleus.

If we assume that neutron capture reactions continued to make the heaviest naturally occurring nuclides, Th^{232}, U^{235}, and U^{238}, out of lower elements like thallium, lead, and bismuth, we find that neutron capture on a slow time scale cannot bridge the gap, and neutron capture on a fast time scale must have been on a very fast time scale indeed.

Figure 2 shows the pertinent nuclear data; the six heaviest stable nuclides are outlined in black on the *Z*, *N* diagram, and the shell numbers $Z = 82$ and $N = 126$ are given special emphasis. Species showing α decay, dropping down two *Z* units and two *N* units, have black triangles in two corners; all half-lives (*10*) for α decay of less than 5 sec are enclosed in circles. The very short α half-lives are the consequence of the very high α-decay energies of all species with $Z \geqslant 84$ and $N \geqslant 128$, since the protons and neutrons have very low binding energies just above the shell numbers 82 and 126. Neutron capture can take matter from the mass-number region below 210 to that above 224 only if neutron addition is on such a rapid time scale that the shell number $N = 126$ is crossed long before β decay raises *Z* to 84. Lack of information about the β-decay properties of very neutron rich species of the elements in this region does not permit a sharp numerical estimate, but it is presumably shorter than the time scale of 8 min for Bi^{215} and 3.0 min for Po^{218}.

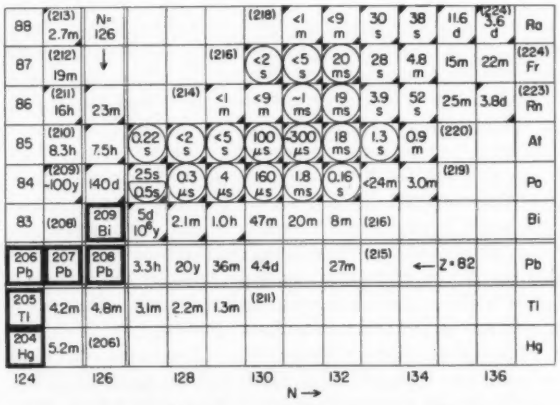

Figure 2. The alpha-decay chute above Bi^{209}. Stable nuclides are shown as black-edged squares with mass number. Half-lives (*10*) are shown for all identified unstable species: μs = microseconds, *ms* = milliseconds, *s* = seconds, *m*, *h*, *d*, and *y* as in Figure 1. Alpha decay is designated by black triangles on the squares; half-lives below 5 sec are encircled. *Ordinate:* atomic number *Z*; *abscissa:* neutron number *N*.

Detailed Nuclidic Abundances

A more quantitative estimate of the time scale for neutron capture in the *r* process can be devised from the intercomparison of elementary abundances so that the relative abundances of various nuclides of different elements can be correlated. This involves estimating the relative amounts of the various elements in the sample of the universe we are considering, a tremendously complicated problem of sampling and chemical analysis.

Suess and Urey (*6*) present such a compendium of analyses as so-called "cosmic abundances." A great deal of data for light elements comes from spectroscopy of the sun and other stars. For the heavier elements, most of the data came from the analysis of meteorites, with suitable weighting of the types iron, sulfide, and stony, with subsidiary arguments to estimate the contribution of the volatile elements (groups VI, VII, and the noble gases). The data for elements of $Z > 30$ came mostly from the solar system, and this is an advantage in the present study, for presumably the history of nucleosynthesis we are tracing is valid for limited a sample. Some unusual stars are known to be rich in heavy elements (barium), and some stars show considerable technetium, which implies massive neutron capture within the half-life of the longest-lived isotope formed in the *s* process, 2.1×10^5-yr Tc^{99}.

Since the publication of the Suess-Urey results (*6*), a large body of work has been done, and many revisions have been proposed; see for instance the paper of Ehmann in this set (*11*). These revisions do not change appreciably the arguments that follow, but they will be of importance in the evaluation of many related problems in nucleosynthesis. For orientation, we list in Table 1 the main features of the Suess-Urey abundances.

Table I. Cosmic (Solar System) Abundances (*6*)

Elements	H	He	C, N, O, Ne
Atom, %	93	7	0.1
Weight, %	75	24	1.2
Elements	Si	Fe	Ga–Bi + Th, U
Atom, %	0.0023	0.0014	7.5×10^{-7}
Weight, %	0.051	0.063	4.6×10^{-6}

The whole body of heavy elements, from gallium through uranium, amounts to a very tiny fraction of the cosmos or of the solar system, in which most of the matter is unburnt hydrogen. The earth as it accumulated from the protoplanetary nebula could not hold the light elements, except for oxygen combined with the metallic elements in the outer part, because of the volatility of H_2, He, CH_4, CO, N_2, H_2O, and Ne[4]. The Suess-Urey abundances, which include corrections for loss of volatile elements from meteorites, give considerable detail over a long sequence of mass number. The data for the region $69 \leqslant A \leqslant 209$ are particularly revealing for the astronuclear processes of transmutation in a very neutron-rich system.

Figure 3 gives the Suess-Urey data (*6*) for abundances of the odd-*A* nuclides from Ga^{69} through Bi^{209}. The ordinate, on logarithmic scale, is abundance *H* (*Häufigkeit*) of the nuclides referred to 10^6 for natural silicon; the abscissa is the mass number *A*. The abundance data for even-*A* nuclides give, in somewhat more complicated form, the same trends and location of peaks.[5] This abundance curve is the first one to give

[4] The hydrogen we have in the oceans presumably came from H_2O liberated from minerals in the crust and mantle.

[5] The even-*A* abundances are higher than odd-*A*, and summation must be made over stable isobars which occur for most *A* values. Little new astronuclear information is carried at the level of the present analysis.

good support to the principle of continuity of abundances, down to the individual mass numbers, which we take as an important principle. This principle is illustrated in the use of the relative abundances of the

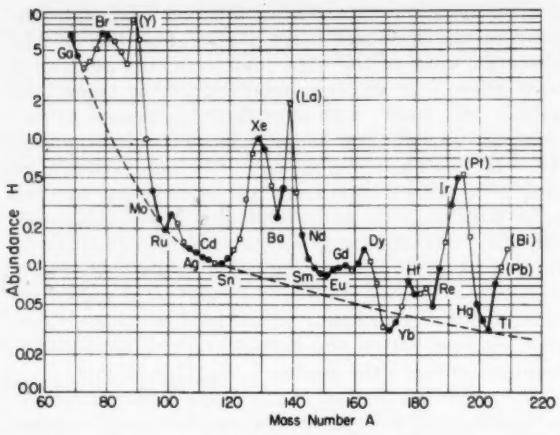

Figure 3. Abundances of the elements, odd-A nuclides from 69–209. Abundance *H* (logarithmic scale) *vs* mass number (6), referred to 10^6 for silicon. Special emphasis for slope is given to data for elements with two stable odd-*A* isotopes.

odd-A isotopes of those elements with two such isotopes to give the slope of the curve. These abundances are marked in Figure 3 as filled circles connected by a heavy line; they serve to give the local value of the slope of the abundance curve. Since isotope fractionation could hardly be appreciable in cosmic processes, these slope data are deemed more secure than corresponding data from the estimates of the relative abundances of different elements, which do fractionate extensively. Current researches on abundances of all the elements may make substantial change in detail (*11*), but except for the region of thallium, lead, and bismuth, they do not challenge the existence of the main structure. In particular, the peaks in the neighborhood of bromine, yttrium, xenon, lanthanum, and iridium-platinum seem well supported.

Abundance Peaks and Shell Numbers

The capture of fast neutrons on a slow time scale (s process) would be expected to lead to an excess of abundance at the neutron shell numbers 50, 82, and 126 because of the low cross-section for their destruction by the (n,γ) process. The sharp peaks at Y^{89} ($N = 50$) and La^{139} ($N = 82$) are considered to mark such a process, reflected also in the high-abundance of the stable even-A species with a closed neutron shell. The areas under these peaks are small, however, compared to those under the three broader peaks. The broken line in Figure 3 gives a rough measure of background, reflecting yields in element synthesis for ordinary species.

It is tempting to ascribe the peaks at bromine, xenon, and platinum to r-process pile-up, in fast neutron capture on a fast time scale, of primary antecedent species with $N^* = 50$, 82, or 126 neutrons (*6, 12*). This would be expected only if the blast of fast neutrons rather suddenly died out, leaving the cosmic material to undergo β decay to the valley of stability without further serious perturbation.

It is useful to define for each peak the number of decays Δ as the difference between the number of neutrons in the species of peak abundance and the shell numbers $N^* = 50$, 82, or 126. For the bromine peak, the Δ value is about 5, the average of 6 for Br^{79} and 4 for Br^{81}, suggesting that these two species came from the initially favored closed-shell species Cu^{79} and Ga^{81}:

$$^{50}_{29}Cu^{79} \xrightarrow{6\beta} {}^{44}_{35}Br^{79} \qquad (7a)$$

$$^{50}_{31}Ga^{81} \xrightarrow{4\beta} {}^{46}_{35}Br^{81} \qquad (7b)$$

These species are even more neutron-rich than primary fission products, of average chain length for β decay of 3 (*13*). The β-decay energies for species of $\Delta \sim 5$ in this mass region can be estimated from β-decay systematics (*1*) as about 9 Mev. This energy corresponds to a half-life of about 0.1 sec for the first step of equations (7*a*) and (7*b*). The half-lives of the subsequent β-decay steps increase as the nuclear potential energy, due to neutron excess, falls.

The Δ value for the xenon peak can be taken as about 7, based on the 82-shell precursors Rh^{127}, Ag^{129}, and In^{131} for stable I^{127}, Xe^{129}, and Xe^{131}, which outline the top of the abundance peak to give Δ values 8, 7, and 5, respectively. The corresponding β-decay energy of the typical first step is about 8 Mev.

The Δ value for the $N^* = 126$ peak topped by Pt^{195} is about 9.5 (10 for Ho^{193} giving rise to Ir^{193}, 9 for Tm^{195} giving rise to Pt^{195}). The decay energy of the typical first decay is about 11 Mev for the $N^* = 126$ peak.

The increase of Δ from 5 to 7 to 9.5 for the successive r process peaks $N^* = 50$, 82, and 126 may be related to the fact that the valley of stability becomes broader as A increases (*1*). It is interesting that the energy of first decay, and thus the probable half-life for the decay, stays about constant. This suggests that the time of recurrence of fast neutron capture is about the same in each of these three regions, and thus, that there was no exhaustion of neutrons as matter in the iron region was built out to mass regions 80, 130, and 194. The r-process peak for $N^* = 50$, plus the Y^{89} s-process peak parasitic to it, stands out about a factor 4.5 above the presumed background (broken line). The peak for $N^* = 82$ plus the La^{139} peak derived from it by s process and the $N^* = 126$ peak also stand out by more than a factor 5 above the background abundances, where shell effects are presumed not to have occurred.

The picture that emerges is that of element synthesis by neutron capture in a tremendous neutron blast. The blast led to steady-state build-up of heavy nuclides from primordial nuclides of A below 69. With the sudden cessation of the neutron blast, the steady-state stream of matter in synthesis, very neutron rich, transformed itself in a sequence of β-decay steps to the heavy elements we now know. The capture cross-sections for fast neutrons (*5*) increase exponentially with A up to $A = 100$. Thus in a stellar core, nuclides like Fe^{56} have a greater capture probability than lighter ones, and nuclei built out to $A = 100$ and above have still greater capture probability.

The question arises, how far can capture go? Figure 2 suggests that the region of high α instability above Bi^{209} is no barrier if neutrons add in repetition times of

0.1–1 sec. Eventually fission will intervene, transforming heavy nuclei into two fragments of near half the mass numbers. But neutron fission of very heavy nuclides that are very neutron rich may not occur until β decay brings the charges up to values well above 92. After the cessation of the neutron blast, β decay will take nuclides of $A > 256$ to the valley of β stability, where spontaneous fission will destroy the species. The abundance curve shows no hump that may easily be correlated with fission (*12*). This may be the result of averaging out of the many kinds of fission that may occur, with recycling of the fast-neutron fission products.

The residue of the steady-state stream that extended above $A = 209$ to the region of $A = 256$, would, after β decay had taken it to the valley of β stability, undergo further α decay. Long half-lives for α decay would hold up much of the matter at Th^{232}, U^{238}, and U^{235}. All of the chain of mass number $4n + 1$ would have decayed to Bi^{209}, as no member exists of half-life comparable with the age of the elements. All members of the other chains $4n$, $4n + 2$, and $4n + 3$ below Th^{232}, U^{238}, and U^{235} would have decayed to the stable species Pb^{208}, Pb^{206}, and Pb^{207}. Problems exist (*11*) about the lead-bismuth abundances compared to expectations for heavy element yieids.

Proton shell numbers $Z^* = 50$ and 82 would not be expected to be marked noticeably in the abundance curve. When β decay occurs in the steady-state stream changing say $^{86}_{49}In^{135}$ to $^{85}_{50}Sn^{135}$, the neutron-capture cross section is expected to fall, along with the β-decay energy, because of the closed proton shell. However, the next β-decay step changes the nuclide to $^{84}_{51}Sb^{135}$, and the proton shell does not occur again. By contrast, on capture of a neutron to give a closed-shell species, say $^{82}_{42}Mo^{124}$, the next β-decay step changes the species to $^{81}_{43}Tc^{124}$, which again captures a neutron to give the closed-shell species $^{82}_{43}Tc^{125}$. This process will occur again and again, with lengthening β half-life, until some member such as $_{50}Sn^{132}$ captures a neutron before β decay, and the chain of neutron addition speeds on to the next shell.

The original interpretation of the fall in neutron capture cross section at closed neutron shells (*12*) has been explored in great detail by Fong (*14*). His treatment involved making a hydrodynamic model of the flow on a Z, N plot, with β decay in competition with neutron capture. Fong assumed a 50-fold fall in cross section for nuclides with $N^* = 50$, 82, or 126. His second parameter is the mean lifetime for neutron capture of non-shell species. The value of 0.1 sec gave good fit to the shapes of the abundance peaks for the r-process. This answer is in accord with the times estimated for β decay of species displaced from the valley of stability by the Δ values derived above, since β decay of these species is in steady-state competition with the neutron capture process forming them.

An alternative explanation, which is probably more realistic, was proposed by the Burbidges, Fowler, and Hoyle (*7*) and independently by Cameron (*8*, *15*). This assumes that neutron capture occurs readily in the process stream until neutron binding energies B^n fall below about 2 Mev, where photo-neutron emission takes place at the same rate as neutron capture. When a species reaches this point in the Z, N diagram, it must wait until β decay takes place, raising Z and thus B^n. Further neutron capture takes place rapidly until the next halt-place occurs. The shell numbers 50, 82, and 126 are well-defined halt points for a number of Z values. The steady-state abundances are relatively high at these halt points. Sudden disappearance of the neutron flux lets this abundance peak be transferred to the same A values in the valley of β stability.

Both explanations provide mechanisms adequate to predict the r-process peaks and their shapes. Interpretation of the regions between the shell peaks will depend on greater knowledge of the dynamic properties and neutron binding of very neutron-rich species, 5 to 10 β decays from stability.

Figure 4 illustrates the path of nucleosynthesis if binding energies set it, using three different sets of estimates for B^n of neutron-rich nuclei, those of the Fowler group (*7*), Cameron (*16*), and Levy (*17*). The

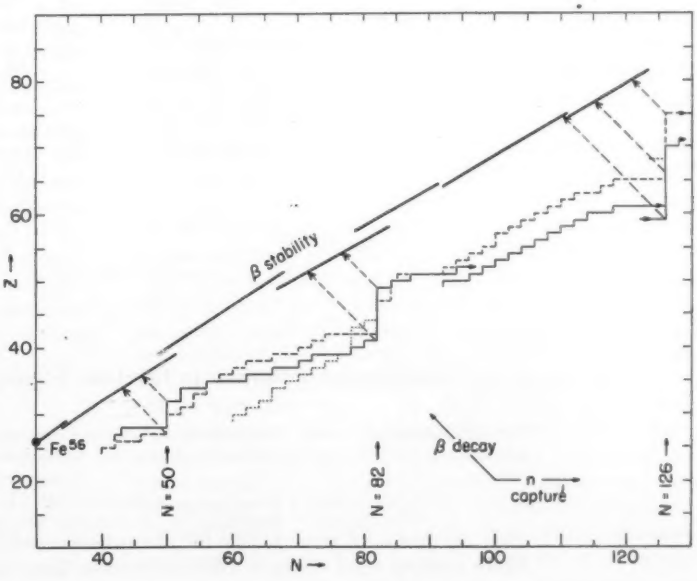

Figure 4. The path of nucleosynthesis in neutron capture on a fast time scale. Postulated is the prompt addition of neutrons until the binding energy B^n falls to 2.0 Mev (*7*, *15*), with halt for β decay. The three capture paths show differences in predictions of B^n; —— (*7*), - - - (*16*), . . . (*17*). After cutoff of neutrons, species in process stream undergo β decay at 135° angle to the valley of stability (*1*). *Ordinate:* atomic number Z; *abscissa:* neutron number N.

heavy tilted lines show the course of the bottom of the valley of stability (*1*); all β-stable odd-A nuclides lie within ± 0.5 Z units of the line corresponding to their Z and N shell classes. The various zig-zag lines show the contours for $B^n = 2.0$ Mev. All three B^n estimates show substantial halt at the abscissas $N^* = 50$, 82, and 126. The three sets differ greatly in the intermediate regions, as much as 10 N (or A) units, in the composition for $B^n = 2.0$. It is possible that increased reliability in abundances will give information about neutron binding of very neutron-rich nuclei.

Conclusion

As shown in Figure 1, isotope abundances of the heavy elements gave indication long ago for neutron capture on a fast time scale in nucleosynthesis. Before nuclear astrophysics provided mechanisms for neutron production in stars, Gamow, Alpher, and Herman (*18, 19*) suggested that all matter started at time zero as a dense neutron fluid, whose expansion and β decay of neutrons to protons led to the creation of all elements by neutron capture while the universe and galaxy were being made. This dramatic hypothesis has serious internal difficulties and is now not deemed necessary nor, in the light of greater knowledge, desirable.

The chemical evidence from detailed abundances (*6–8, 15*) indicates that a large fraction of the synthesis of elements above the iron region was by fast neutron capture on a very fast time scale, with sudden cut off of neutrons (*r*-process). Subsidiary processes (*p* process, *s* process, fission, and α decay) gave the neutron-poor species and made modifications of the abundances of other species. Supernova explosions give an attractive mechanism for the *r*-process (*7, 8*).

Our sun cannot make heavy elements, or even the medium weight elements carbon through iron-nickel known to be present (*6–8*). Thus it is argued that the supernova cycle preceded the aggregation of the matter in our solar system (*7, 8, 20*). From present knowledge of astrophysics, the universe is old enough and has had enough mixing, that the heavy elements of the solar system may have come from many supernovas, and indeed, all of the elements around us may have gone through many stellar cycles.

An interesting situation now exists, where analytical chemistry, especially in meteorites, and nuclear chemistry both have a strong bearing on astrophysics and cosmology. It is the author's sincere hope that the new alliance (cosmochemistry) will bring new perceptions for the chemistry of Creation (*21*).

Acknowledgments

The author is grateful to Professor Harold C. Urey and Professor Hans E. Suess of the University of California at La Jolla, to Professor William A. Fowler of the California Institute of Technology, and to Dr. Alistair G. W. Cameron of Atomic Energy of Canada, Ltd., Chalk River, for encouragement and valued advice and criticism. This work was supported by the U. S. Atomic Energy Commission.

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Truman P. Kohman
Carnegie Institute of Technology
Pittsburgh, Pennsylvania

Chronology of Nucleosynthesis and Extinct Natural Radioactivity

The simple question "How old are the elements?", often asked (and frequently answered!) in the past, must on the basis of present knowledge be replaced by a more meaningful one, "When were the elements of the solar system synthesized?" It is now evident that the shuffling of nucleons to form the nuclei of the elements as they now exist must have been a complicated process involving a variety of mechanisms operating at several or many different times, and that a particular sample of matter may have had an entirely different history than a sample in another part of the universe. We limit our inquiry for the present, accordingly, to the "local" sample of elements, about which we have by far the most information, and in which we have the greatest personal interest. At the same time we broaden the question to concern the entire span of time within which nucleosynthesis of "our" elements occurred, as well as to include something about the time dependence of the synthesis within that span.

General Ideas on the Time of Nucleosynthesis

Until recently it was supposed that all of the elements were created much in their present form in one violent event, referred to as "nucleogenesis," which may have been intimately connected with the initial stage of the expanding universe (*1*, *2*). According to this idea all of the atoms would have the same definite "age," which might be determined either from the mutual recession time of the extragalactic nebulae or from the residual content of long-lived natural radionuclides; the lower limits of this "age" would be given by the ages of any bodies or substances in the universe. Serious difficulties with the development of this idea and recent developments in astrophysics have subsequently led to general acceptance of the alternate idea that "nucleosynthesis," or the building up of the heavier atomic nuclei from light particles, proceeds mainly in stellar interiors with redistribution of the matter mainly through supernova explosions (*3–7*). This idea is compatible both with a single nucleogenic event coincident with "cosmogenesis" and with "continuous creation" of matter in a steady-state expanding universe. Nucleosynthesis thus follows but is not directly connected with "nucleogenesis," the initial creation of the elementary particles and hence of matter itself.

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Among the hundreds of nuclides composing "our" matter, those which are most useful for giving information about the time of its formation are those which are radioactive with half-lives sufficiently short for appreciable decay to have occurred during or subsequent to the formation period. Of the score of "primary" natural radionuclides, only four, U^{235} [half-life $(7.0 \pm 0.1) \times 10^8$ y], K^{40} [$(1.27 \pm 0.01) \times 10^9$ y], U^{238} [$(4.51 \pm 0.01) \times 10^9$ y], and Th^{232} [$(1.42 \pm 0.02) \times 10^{10}$ y], are sufficiently short-lived. For all others, which have half-lives greater than 2×10^{10} years, depletion by spontaneous decay has been relatively unimportant.

For quantitative conclusions, it is necessary to have information about the mechanism of formation of the nuclides. Until recently it has also been supposed that the initial relative abundances of the uranium isotopes would follow the same regularities observed among the stable elements, with the odd-A U^{235} being produced in smaller quantities than its even-A isotope U^{238}. This assumption and the present value of the U^{235}/U^{238} ratio $[(137.8 \pm 0.2)^{-1} = 0.00726 \pm 0.00001]$ then set an upper limit on the "age of the elements" of ~ 6 *Æ* (*Æ* = aeon = 10^9 years) (*8*). A similar limit, <10 *Æ*, results from the reasonable assumption that the initial isotopic abundance of the odd-odd nuclide K^{40} was $<2.5\%$ of all potassium isotopes (*9*). More sophisticated heavy-element abundance systematics, but retention of the single-age idea, gave for this 6.25 ± 0.5 *Æ* (*10*). These values or limits are definitely higher than the values ~ 4.5 *Æ* given by several radioactive dating methods for the "age of the meteorites," a concept which also requires more precise formulation, but which is probably not much less than the "age of the solar system," of which system the meteorites are, and probably always have been, a part (*11*).

More precise dating of element formation could be obtained from information about the decay of shorter-lived radionuclides, the primordial supply of which would now be completely extinct, but whose products might be found in the oldest bodies available for isotopic analysis, the meteorites (*12*). The amount of radiogenic decay product would allow an estimation of the interval between nucleosynthesis and the governing chemical fractionation in the formation of the meteorite. Addition of this interval to the "age of the meteorite" would give the "age of the elements." All searches for such cases, most significantly for radiogenic Xe^{129} from the now-extinct I^{129}, were until very recently unsuccessful (*13*, *14*); the interval in question was concluded to be >0.4 *Æ*. Since the difference between the "age of the solar system" and the "age of the elements" appeared to be at least 1 *Æ*, it appeared increasingly

unlikely that any case of "extinct natural radioactivity" would be found (*15*).

According to the view that the nuclei heavier than helium have been synthesized in stars and ejected into interstellar space, nucleosynthesis has been occurring in our galaxy continuously throughout most of its lifetime, even down to the present time. The synthesis of the elements of the solar system, however, presumably ceased as its matter condensed into a volume of space too small for sweeping up appreciable amounts of interstellar gas and dust. B_2FH [Burbidge, Burbidge, Fowler, and Hoyle (*3*)] described eight different processes by which complex nuclei seem to be synthesized in stars from primeval hydrogen. Of particular interest for the time scale is the "r-process," or rapid build-up of nuclei by neutron capture in exploding supernovae; this is the only process by which the heaviest nuclides (including the above mentioned U^{235}, U^{238}, and Th^{232}) can be produced, and for it relative production rates can be calculated with some feeling of confidence (*3*, *16*).

B_2FH (*3*) suggested as an approximation a model in which the heavy nuclei were produced at constant rates with uniform mixing for a definite period of time. In a more recent paper, Fowler and Hoyle (*16*) have generalized this model to include variation in the production rates, which have most likely been decreasing steadily as more and more matter of the galaxy is tied in non-exploding stars. For simplicity they assumed an exponentially decaying rate of "stellar activity" responsible for nucleosynthesis. They also pointed out that the r-process would not start immediately with the formation of stars, but would be delayed by the time required for the Type I supernova stars, in which the r-process chiefly occurs, to evolve to the exploding stage.

Figure 1 shows schematically the course of events during and subsequent to the synthesis of our

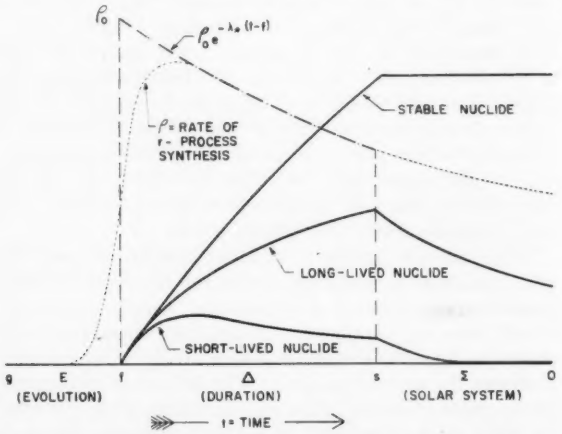

Figure 1. Schematic course of heavy-element nucleosynthesis in the galaxy, showing the time dependence of r-process nucleosynthesis and of the amounts of various kinds of r-process nuclides.

elements. The present time is represented by $t = 0$, and other specific times are indicated by small Latin letters: g = origin of galaxy (nucleogenesis?); f = beginning of formation of r-process nuclides; s = termination of nucleosynthesis for elements of the solar system (origin of solar system?). Specific intervals are represented by capital Greek letters: $\mathbf{E} = f - g$ = average time for evolution of supernova stars; $\Delta = s - f$ = duration of nucleosynthesis for heavy elements of the solar system; $\Sigma = 0 - s$ = evolution of solar system without further nucleosynthesis. The dotted line shows the time dependence of supernova explosions and hence of r-process nucleosynthesis. The dashed line shows a simplified representation of this in which the rate is assumed to decay exponentially since the mean time of beginning with the decay constant $\lambda_\star$. It is assumed that the production rate, ρ, of any particular nuclide is always proportional to the general rate of nucleosynthesis after time f. The solid curves show the time dependence of a stable nuclide, a long-lived nuclide (such as U^{238}) and a moderately short-lived nuclide (such as U^{235}) which are produced only by r-processes.

Our problem, mathematically stated, is the evaluation of the three quantities Σ, Δ and $\lambda_\star$. This requires, in a general way, three pieces of information.

Termination Date of Nucleosynthesis

According to the method proposed by Brown (*12*) the time subsequent to nucleosynthesis, Σ, could be obtained as a sum of two intervals, M = age of meteorites, and Ξ = decay interval for an extinct natural radionuclide, provided a case of extinct natural radioactivity could be found.

Decay Interval for Extinct I^{129}

Very recently the first positive observation of extinct natural radioactivity has been made. Reynolds (*17*) has detected an excess of Xe^{129} in the minute trace of xenon present in the chondrite meteorite Richardton, and has attributed it to the presence of a trace of I^{129} remaining at the time the bulk of the primordial xenon was lost from the parent body of the meteorite and it became able to retain radiogenic xenon. The observation has been confirmed in other laboratories (*18*, *19*), and radiogenic Xe^{129} has been found in other meteorites (*20–23*).

Using the single-nucleogenesis hypothesis and making reasonable assumptions about the original amount of I^{129}, Reynolds (*17*) calculated a time of 0.35 ± 0.06 *Æ* for the decay of I^{129}. However, if nucleosynthesis is a process of extended duration, the time calculated by Reynolds is an *upper limit* for the interval between the *termination* of the nucleosynthesis for the elements of the meteorite and the *beginning* of its retention of the noble gas xenon (*24*). Fowler and Hoyle (*16*) state that Reynolds' results imply that this interval is 2×10^8 years, which yields a reasonable time, $\sim 10^{10}$ years, for the duration of nucleosynthesis at a uniform rate (*24*).

Since the amount of I^{129} present after nucleosynthesis depends critically on the interval (Ξ) between the termination of its production ($t = s$) and the beginning of the retention of its daughter ($t = s + \Xi = x$), but is much less sensitive to the duration of nucleosynthesis (Δ), it is more reasonable to assume the latter and to calculate the former. If we let ρ_9 and ρ_7 be the production rates of I^{129} and I^{127}, N_9 and N_7 be their numbers of atoms, N_x be the number of radiogenic Xe^{129} atoms produced subsequently to $t = x$, and λ_9 be the disintegration constant of I^{129} [= ln 2/(1.64 ±

0.1) $\times 10^7$ y], then

$$\left(\frac{N_X}{N_7}\right)_{t=0} = \left(\frac{N_9}{N_7}\right)_{t=\Xi} = \frac{\rho_9}{\rho_7}\frac{\lambda_\star}{\lambda_9}\left(e^{\lambda_\star \Delta} - 1\right)^{-1} e^{-\lambda_9 \Xi} \quad (1)$$

This allows Ξ to be calculated as a function of the experimental quantity $(N_X/N_7)_{t=0}$ and assumed values of ρ_9/ρ_7, $\lambda_\star$, and Δ.

Reynolds (*17*) determined that Richardton contained a radiogenic Xe^{129} content of 1.3×10^{-10} cm^3 g^{-1} and assumed an I^{127} content of 1 ppm, which gives $(N_X/N_7)_{t=0} = 7.4 \times 10^{-7}$. Adopting his assumption $\rho_9/\rho_7 = 1$, equation (1) yields for any reasonable combination of Δ in the range 2–10 *Æ* and $\lambda_\star$ in the range 0–0.3 *Æ*$^{-1}$ a value of Ξ not far from 0.20 *Æ*, instead of the value calculated by Reynolds for the case $\Delta = 0$ [$\lambda_\star = \infty$]. Goles and Anders (*25*) have subsequently measured the iodine content of Richardton by neutron activation analysis, obtaining 47 ± 14 ppb as an average. This leads to $(N_X/N_7)_{t=0} \approx 1.6 \times 10^{-5}$ and $\Xi \approx 0.12$ *Æ*. Reynolds (*21*) has developed a clever method of measuring $(N_X/N_7)_{t=0}$ directly, obtaining $\sim 1.0 \times 10^{-5}$ for Richardton and essentially the same value for Ξ. Quite similar values of Ξ have subsequently been obtained for two other meteorites (Murray, Indarch) in which Reynolds (*20, 21*) has found even larger contents of radiogenic Xe^{129} and Goles and Anders (*25*) have determined the correspondingly higher iodine contents. On the other hand, the meteorites containing no detectable radiogenic Xe^{129} have values of $\Xi > 0.2$ *Æ* (*25*). We may thus take $\Xi = 0.2 \pm 0.1$ *Æ* as the best present estimate of the average value for meteorites.

Age of Meteorites

Radioactive age determinations date chemical separations or fractionations of the parent and daughter elements, and for different pairs of genetically related elements the effective or last time of separation may not be the same. For the xenon-iodine separation, the effective time is that at which the solid matter had cooled sufficiently to retain xenon. The effective fractionation of non-volatile elements could have occurred at higher temperatures and hence somewhat earlier. Thus, the "age" of the meteorites given by the uranium, thorium-lead, and rubidium-strontium dating methods as 4.6 ± 0.1 *Æ* (*26–29*) is an *upper limit* to the time (M) we are seeking here.

The potassium-argon ages of meteorites are almost always somewhat lower, ranging for chondrites from ~2 to ~4.3 *Æ*, as a result of argon losses. Goles, *et al.* (*30*) have given a detailed discussion of losses of argon from stone meteorites by diffusion, concluding that probably in all cases some argon losses have been occurring continuously. Thus the apparent argon ages must be corrected upward to give the actual time, since most of the argon was retained. In the case of Richardton, apparent K-Ar dates of 4.2 ± 0.1 *Æ* (*31*) and 4.4 ± 0.2 *Æ* (*20*) have been obtained, and even these dates must be regarded as *lower limits* on M.

Considering both of these limits, we may tentatively take M = 4.5 ± 0.1 *Æ* as the time since Xe^{129} retention.

Simple addition gives $\Sigma = \Xi + M = 4.7 \pm 0.2$ *Æ* for the time since the last nucleosynthesis of the elements of the solar system. The same figure was adopted by Fowler and Hoyle (*16*) on similar and other grounds. This is the first of the three quantities to be evaluated.

Duration of Nucleosynthesis

Because of the long spans of time involved, only comparatively long-lived radionuclides can be of use in calculating the other two quantities, Δ and $\lambda_\star$. Fortunately, from the cosmic abundances of the heavy r-process nuclides U^{235}, U^{238}, and Th^{232}, two independent ratios can be obtained, providing for the evaluation of two additional parameters.

Calculations of Fowler and Hoyle

B_2FH (*3*) calculated the relative production rates of U^{235}, U^{238}, Th^{232}, and other heavy nuclides in supernovae in the r-process. They also showed that if these rates were constant during nucleosynthesis, ($\lambda_\star = 0$), it was possible to calculate the duration of nucleosynthesis from the present U^{235}/U^{238} ratio provided the time of the beginning or of the end were known. That was not the case in 1957. A similar situation prevailed for the U^{238}-Th^{232} pair, but in 1957 the Th^{232}/U^{238} ratio was not well enough known.

In their recent paper, Fowler and Hoyle (*16*) have re-estimated the relative production rates of these three nuclides, obtaining $\rho(U^{235})/\rho(U^{238}) = 1.65 \pm 0.15$ and $\rho(Th^{232})/\rho(U^{238}) = 1.65 \pm 0.15$. They have also pointed out that nucleosynthesis has probably not been constant, so that even if one end of the nucleosynthesis period is known independently, the duration cannot be determined uniquely from the present U^{235}/U^{238} ratio without knowing the time dependence of nucleosynthesis. However, the two present-day ratios U^{235}/U^{238} and Th^{232}/U^{238} together with the independently fixed termination date will allow simultaneous determination of the starting date and a parameter characterizing the time-dependence of the production rates.

Considering various data, but principally the Th/U ratio in chondrites, Fowler and Hoyle selected 3.80 ± 0.3 as the best value of Th^{232}/U^{238} today. Using this, $U^{235}/U^{238} = 0.00723$ today, and 4.7 *Æ* for the age of the solar system, they derived 11.6 *Æ* for the time since the beginning of nucleosynthesis and a rate of exponential decay of stellar activity such as to yield a present-day galactic nucleosynthesis rate 26% of that 15 *Æ* ago. The latter figure is the corresponding age of the galaxy, considering an induction period of E = 3.3 ± 0.5 *Æ* in the r-process, this being the mean lifetime of Type I supernova stars before eruption. In our symbolism, their solution implies $\lambda_\star = 0.090$ *Æ*$^{-1}$.

Fowler and Hoyle indicated that the time of beginning of nucleosynthesis, and hence the age of the galaxy, on this model is uncertain by several aeons, being rather sensitive to the values of Th^{232}/U^{238} and the production rate ratios used. However, the general indication that the age of the galaxy is much greater than that of the sun and its family is in line with other astronomical evidence. Some stars seem to have required 10 *Æ* or more to have developed to their present status (*32–35*). The expansion-time of the universe, $^2/_3$ of the reciprocal of the Hubble constant for nebular recession, has been estimated by Sandage as probably lying in the range 7 to 13 *Æ* (*36*).

Fowler and Hoyle also considered in their paper (*16*) a radically different production model for the source of our elements, according to which there is a substantial interchange of matter between nebulae through intergalactic space. This does not yield a definite time for the beginning of nucleosynthesis or the age of our galaxy, but yields instead an independent value of the Hubble constant, whose reciprocal turned out to be ~11 Æ, again subject to uncertainty of several aeons because of uncertainties in the data, but agreeing in order of magnitude with that derived from nebular recession. A choice between the two models did not seem to be possible, but Fowler and Hoyle pointed out that the latter model avoids a serious quantitative discrepancy of the more conventional model in the cosmic abundances in the heavy r-process nuclides.

Calculations of the Present Author

Following the first announcement of Reynolds' finding of radiogenic Xe^{129}, the present writer undertook calculations on the chronology of nucleosynthesis, with results similar to those of Fowler and Hoyle. These calculations, the details of which will be published elsewhere (*37*) have been carried out in such a way as to permit the ready evaluation of the effects of different choices of the input data $(N_5/N_8)_0$, $(N_2/N_8)_0$, ρ_5/ρ_8, ρ_2/ρ_8, and Σ on the output values Δ and $\lambda_\star$. Here N_5, N_8, and N_2 refer to the relative atomic abundances of U^{235}, U^{238}, and Th^{232}; ρ_5, ρ_8, and ρ_2 refer to their relative production rates in supernovae; also, λ_5, λ_8, and λ_2 will refer to their radioactive decay constants.

Solution of the differential equation

$$\frac{dN_i}{dt} = \rho_{i,0}\, e^{-\lambda_\star(t-f)} - \lambda_i N_i \qquad (2)$$

leads to expressions for the atomic ratios N_5/N_8 and N_2/N_8 as functions of Σ, Δ, and $\lambda_\star$. Figure 2 shows a typical plot of the results, in this case for $\lambda_\star = 0$ and the Fowler-Hoyle values of ρ_5/ρ_8 and ρ_2/ρ_8 given above. The heavy dashed line shows how the ratios

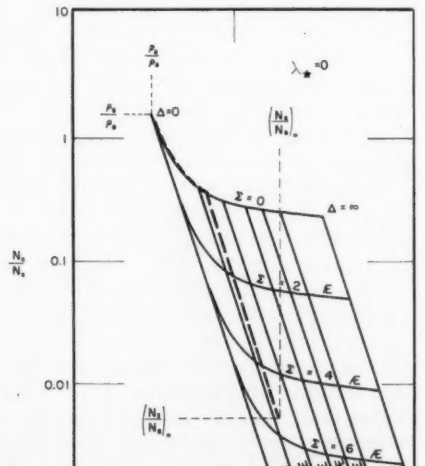

Figure 2. Variation of relative quantities of U^{235}, U^{238}, and Th^{232} during and subsequent to nucleosynthesis, as functions of Δ (duration) and Σ (subsequent time), with constant rates of nucleosynthesis ($\lambda_\star = 0$).

N_5/N_8 and N_2/N_2 will vary together, in the galaxy as a whole along the top curve ($\Sigma = 0$), and locally along a diagonal corresponding to a given value of Δ after removal from the mixing region, terminating at a point corresponding to the appropriate value of Σ. From the present ratios $(N_5/N_8)_0$ and $(N_2/N_8)_0$, one could read off Δ and Σ by interpolation. This could be repeated for different values of $\lambda_\star$ until the proper value of Σ was obtained, the corresponding values of Δ and $\lambda_\star$ being the proper ones.

Since estimates of ρ_5/ρ_8 and ρ_2/ρ_8 are subject to variation, it is better to use a plot of

$$\frac{(N_5/N_8)_0}{\rho_5/\rho_8} \text{ versus } \frac{(N_2/N_8)_0}{\rho_2/\rho_8}$$

Moreover, we are only interested in the region of the plot containing the likely values of these ratios. Figure 3 shows such a "magnified" plot, in this example for the case $\lambda_\star = 0.1$ Æ$^{-1}$. For a selected pair of ordinate

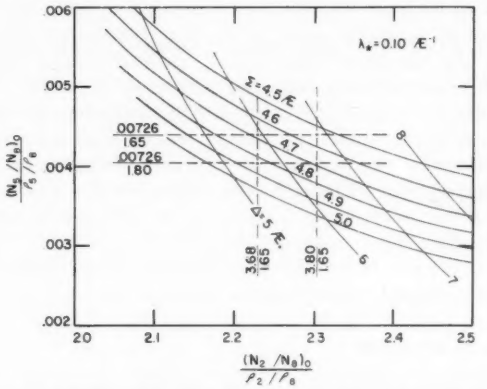

Figure 3. Variation of U^{235}, U^{238}, and Th^{232} abundance ratios relative to production-rate ratios for various combinations of Δ (duration of nucleosynthesis) and Σ (subsequent time), with moderately decreasing rates of nucleosynthesis ($\lambda_\star = 0.10$ Æ$^{-1}$).

and abscissa values, the corresponding values of Δ and Σ for the given value of $\lambda_\star$ can be read off. This is done for several values of $\lambda_\star$, and a plot of Σ versus $\lambda_\star$ is made. Since Σ is known, $\lambda_\star$ is thereby determined. From a corresponding plot of Δ versus $\lambda_\star$, Δ is similarly determined from the now-known value of $\lambda_\star$.

Outside of the production rate ratios, the least certain datum is $(N_2/N_8)_0$. Fortunately, a survey of the Th/U ratio in stone meteorites has recently been made by König and Wänke (*38*), based in part on their own new uranium analyses. Five chondrites have been analyzed both for thorium and for uranium by neutron activation analysis, the ratio being considerably more constant than the individual element concentrations. The five values (3.02, 3.12, 3.66, 3.76, and 4.22) average 3.56 ± 0.22. Dividing this by $U^{238}/U = 0.99274$, and multiplying by the ratio of atomic weights of uranium to thorium, yields $(N_2/N_8)_0 = 3.68 \pm 0.22$.

Because of the uncertainty in ρ_5/ρ_8, ρ_2/ρ_8, and $(N_2/N_8)_0$, and in order to show the sensitivity to the exact values used, four solutions have been obtained, corresponding to two different values each of $(N_2/N_8)_0$ and of ρ_5/ρ_8. Table 1 lists the input and output values. Case *A* corresponds to that used by Fowler and Hoyle [the result for $\lambda_\star$ differs from theirs (0.09 Æ$^{-1}$), possibly due to their use of different λ_5, λ_2, and $(N_5/N_8)_0$].

Table 1. Derived values of $\lambda_\star$ and Δ

Case	Input values					Output values	
	ρ_5/ρ_8	ρ_2/ρ_8	$(N_5/N_8)_0$	$(N_2/N_8)_0$	$\Sigma(Æ)$	$\lambda_\star(Æ^{-1})$	$\Delta(Æ)$
A	1.65	1.65	0.00726	3.80	4.70	0.01_5	7.2
B	1.65	1.65	0.00726	3.68	4.70	0.08_5	6.0
C	1.80	1.65	0.00726	3.80	4.70	0.09_5	6.6
D	1.80	1.65	0.00726	3.68	4.70	0.19	5.4

Case *B* uses the value of $(N_2/N_8)_0$ derived from the latest analyses of chondrites. The values of $\lambda_\star$ in Cases *A* and *B* correspond respectively to stellar activity survival factors in time $\Gamma = E + \Delta + \Sigma$ (with $E = 3.3$ Æ) of 0.80 and 0.30 (Fowler and Hoyle obtained 0.26 for the first case). Cases *C* and *D* differ from *A* and *B* only in the use of a value of one of the production rate ratios differing from its "best" value by one standard deviation. The corresponding survival factors are 0.25 and 0.08, respectively.

It is evident that both $\lambda_\star$ and Δ are quite sensitive to each of the quantities ρ_5/ρ_8, ρ_2/ρ_8, and $(N_2/N_8)_0$. For the present, it can be concluded that the data are consistent with a "long" time scale for nucleosynthesis, ending 4.7 Æ ago and starting several aeons previously, and inconsistent with a "sudden" nucleogenesis-nucleosynthesis. The data are insufficiently precise to decide conclusively whether $\lambda_\star$ differs from zero, but they are consistent with a moderate finite value. Since Case *B* probably represents the best present set of input values, the best present estimate of $\lambda_\star$ is $\sim 0.1\ Æ^{-1}$ and of Δ is ~ 6 Æ.

Radiogenic Heat Subsequent to Nucleosynthesis

From the known present abundances of the primary natural radionuclides it is possible to calculate their abundances at any time in the past, and consequently the total rate of generation of radiogenic heat. Whereas in the earth at the present time the heat contributions of the U^{238} and Th^{232} families and K^{40} are of comparable magnitude, K^{40} has played a dominant role throughout past geologic history. If the average concentrations of potassium, uranium, and thorium in chondrites are representative of the solar system, K^{40} has been dominant, and the heat production rate from all known radioactive sources was about 10 times as great 4.7 Æ ago as it is now. The question has frequently been raised as to whether one or more now-extinct nuclides have been important early in the history of the solar system. Urey (*39*) in particular suggested that the present characteristics of the meteorites and planets required such a source of supplementary heat. This, however, was regarded (*15*) as quite unlikely on the basis of single-event nucleogenesis, because of the relatively long time between such an event and the condensations to form the minor bodies of the solar system.

Very recently Fish, *et al.* (*40*) have again concluded from observations on the physical and chemical characteristics of meteorites that rather intense heating of their parent bodies must have taken place during their development. The heat supply must have been many times that of K^{40} plus all other known sources, and they deduced that now-extinct radioactivity must have been responsible. They assumed a single nucleosynthetic event and an almost immediate formation of the meteorite parent bodies. If the intervening interval were as short as ~ 5 M y, Al^{26} and Fe^{60} could have supplied the necessary heat. If some unknown nuclide had suitable characteristics, the interval could have been considerably longer.

The presence of now-extinct radioactive heaters in the parent bodies of the meteorites is much more reasonable on the basis of long-term nucleosynthesis than on the basis of a single nucleosynthetic event. However, the relative quantity of a short-lived nuclide at the termination of a long steady nucleosynthesis is very much less than that present immediately after a very short nucleosynthetic event. Accordingly it is doubtful whether even on this model any now-extinct nuclide could have produced very great amounts of heat except for a trivially short period after the termination of its synthesis.

Continuous Nucleosynthesis

Having a fairly definite time for the cessation of nucleosynthesis and an approximate knowledge of its duration, it is possible to estimate the amounts of all radionuclides present at all times following the cessation. Calculations were actually made for the parameters $\Sigma = 4.70$ Æ, $\lambda_\star = 0$, and $\Delta = 7.3$ Æ, but the results are relatively insensitive to the latter two parameters. The details will be given elsewhere (*37*).

Estimations of the relative production rates of both stable and radionuclides are necessary. For stable nuclides, the present cosmic abundances as given by Suess and Urey (*41*), which for non-volatile elements are derived mainly from analyses of chondrites [see also Ehmann (*42*)], were used. For primary natural radionuclides, the present weight fractions in chondrites were corrected for radioactive decay both subsequent to and during the production period. For extinct radionuclides with $Z \leqslant 83$, the production rates were estimated from the cosmic abundances of near-by stable nuclides, considering patterns of isotopic abundances, probable production mechanisms, and in each case the present abundance of the stable product, which must not be exceeded. For heavy r-process nuclides, the relative production rates given by B_2FH (*3*) were used, combining with each long-lived nuclide all shorter-lived antecedents, making certain corrections and suitable normalizations.

All known radionuclides with half-lives from 10^5 y to 10^{11} y were considered. For each, the energy of disintegration, including that of all shorter-lived descendants but excluding neutrinos, was calculated.

The relative numbers of atoms present at the end of nucleosynthesis ($t = s$) were then calculated, and for the radionuclides the initial specific heat production rates in chondritic matter were computed. These values were then computed for subsequent times by simple decay corrections. Addition of the contributions of all nuclides at a given time gave the total heat generation at that time.

Figure 4 is a plot of the various contributions to radiogenic heat for the entire lifetime of the solar system. Figure 5 is a plot for the first 35 million years after the cessation of continuous nucleosynthesis. It is seen from the figures that, except for very short-lived nuclides not considered ($H < 10^5$ y), only Al^{26} and Fe^{60} (with its Co^{60m} and Co^{60g} daughters) exceed K^{40} in heat production. The total contribution from

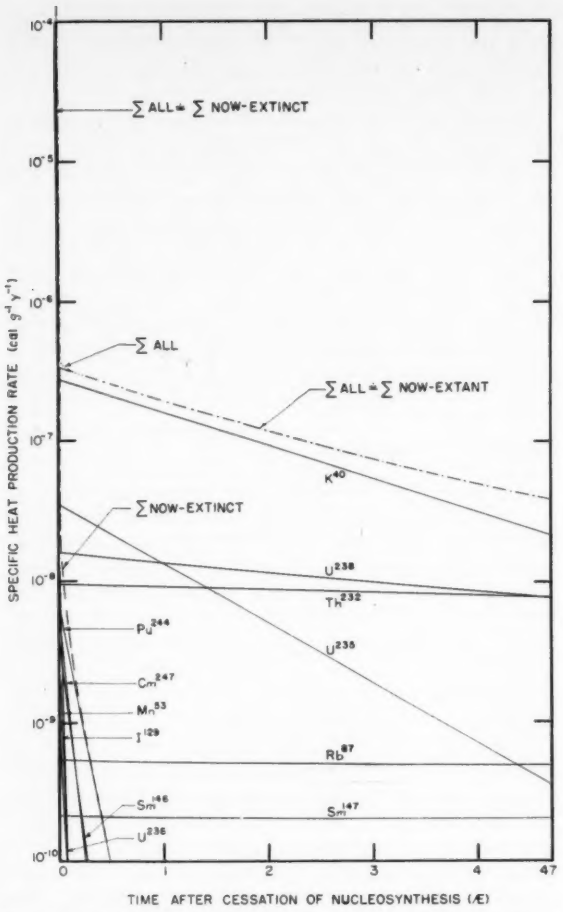

Figure 4. Contributions to radiogenic heat in chondrite-like matter of various nuclides with half-lives $\geq 10^7$ years, with totals from now-extant, now-extinct, and all nuclides, at times from the cessation of continuous nucleosynthesis until the present.

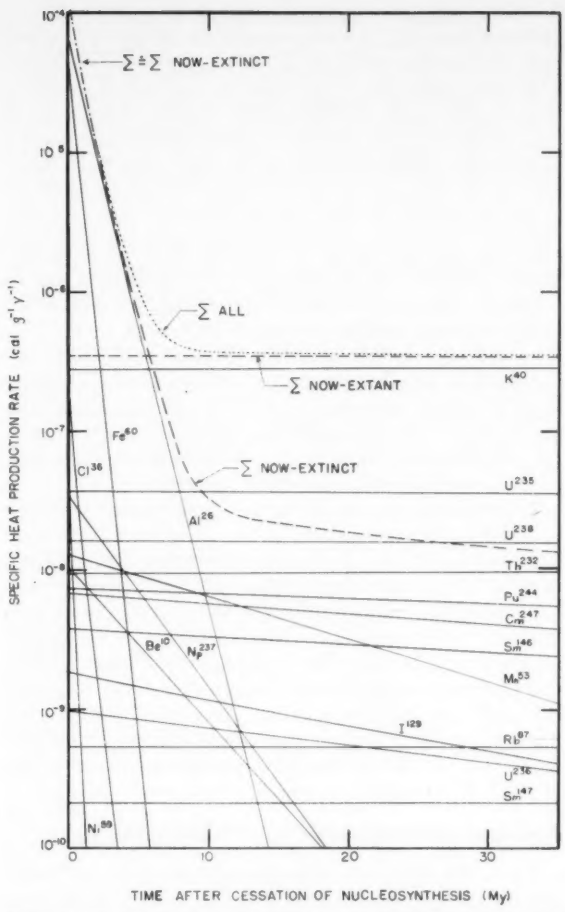

Figure 5. Contributions to radiogenic heat in chondrite-like matter of various nuclides with half-lives $\geq 10^6$ years, with totals for now-extant, now-extinct, and all nuclides, at times up to 35 million years after the cessation of continuous nucleosynthesis.

now-extinct heaters falls to equality with the total from the now-extant heaters at 6 M y and is relatively unimportant after 10 My = 0.01 *Æ*.

According to Fish, *et al.* (*40*), the meteorite parent bodies must have been heated to ~2000 or 3000°K. With a heat capacity of ~0.2 cal deg^{-1} g^{-1} and a heat of fusion of ~100 cal g^{-1}, an integral heat generation of at least ~1000 cal g^{-1} would be required in a time short enough to avoid much loss from moderately small bodies. A specific heat production of ~10^{-3} cal g^{-1} y^{-1} acting for ~1 My or the equivalent would be needed. It is evident that no such possibility exists on the model of steady production terminating before the condensation of bodies in the solar system. Although a hypothetical nuclide with half-life ~10^8 y and large production rate and disintegration energy could modify this conclusion, it is very unlikely that such a now-unknown nuclide exists. Moreover, such a long-lived heat source would be incompatible with the observed retention of Xe^{129} in meteorites.

Single-Event Nucleosynthesis

The relative numbers of atoms of radionuclides and their heat production rates were also calculated for the case of a single nucleogenetic-nucleosynthetic event, as considered by Fish, *et al.* (*40*). The relative numbers of atoms were taken to be directly proportional to the above-derived production rates, even though there would certainly be differences in such a case. The present abundances of the primary natural radionuclides were extrapolated back to 6.5 *Æ* ago as required by the present U^{235}/U^{238} abundance and production-rate ratios. The results are shown in Figure 6, which is similar to Figure 5 of Fish, *et al.*

The heat produced by now-extinct nuclides on this model exceeds that by now-extant nuclides for a longer time, ~65 My. However, with such a source of our elements, a considerably longer time would undoubtedly be required for condensation. Furthermore, the event would have to have occurred close to 6.5 *Æ* ago, long before the heat is needed, ~4.7 *Æ* ago.

"Mixed" Nucleosynthesis

Is it possible to construct a model of nucleosynthesis which would account for both the relatively intense but short-lived heat source indicated by the meteorites and the long production period required by the relative abundances of U^{235}, U^{238}, and Th^{232}? Suppose the bulk of our elements were produced by a more-or-less steady-state process of several aeons duration ending ~4.7 *Æ* ago, and at that time a small fraction, φ,

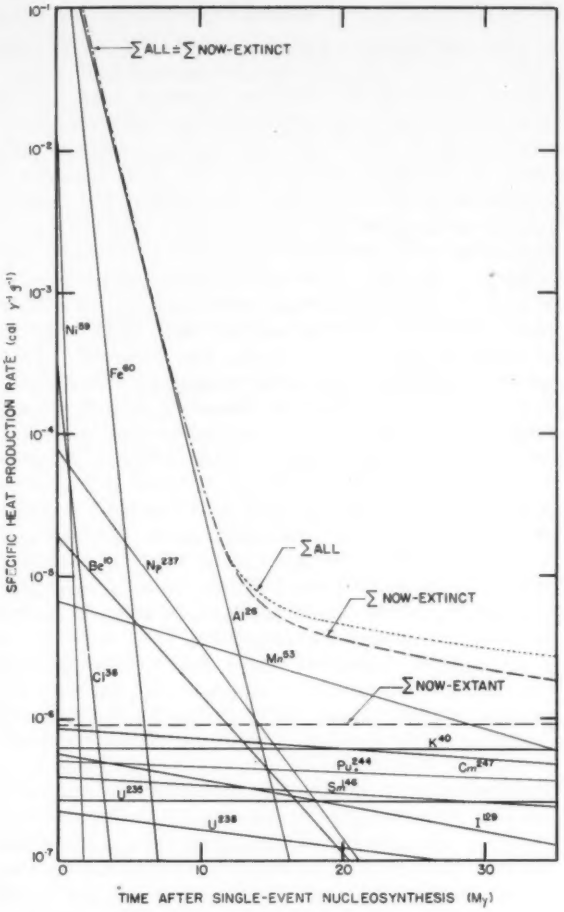

Figure 6. Contributions to radiogenic heat in chondrite-like matter of various nuclides with half-lives $\geqslant 10^5$ years, with totals for now-extant, now-extinct, and all nuclides, at times up to 35 million years after a single nucleo synthetic event occurring 6.5 Æ ago.

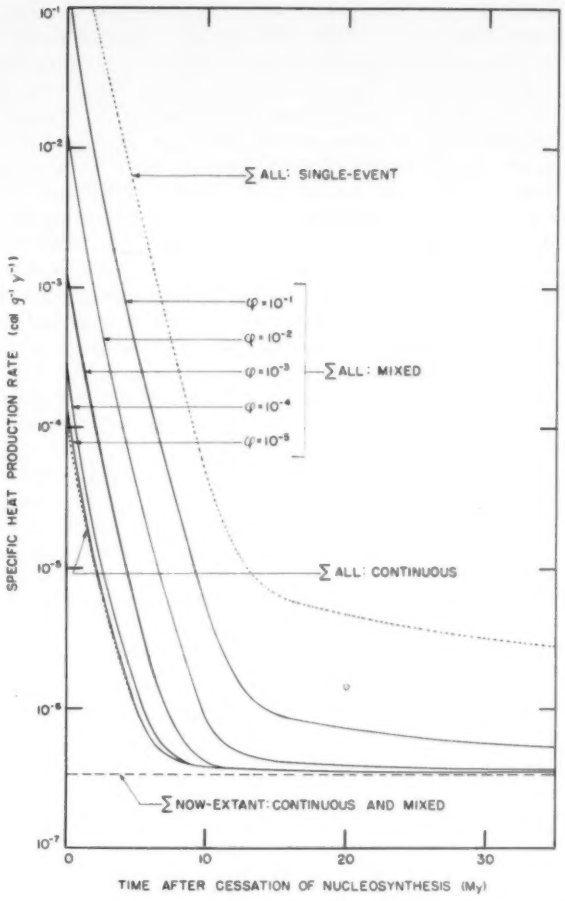

Figure 7. Radiogenic heat production in chondrite-like matter from all nuclides with half-lives $\geqslant 10^5$ years, for mixed nucleosynthesis in which a fraction (1-φ) of the matter was produced by continuous nucleosynthesis and a fraction φ was produced by a single nucleosynthetic event at the termination of continuous nucleosynthesis 4.7 Æ ago.

of matter from a single supernova was added, just before or actually during the condensation of the proto-bodies of the solar system.

The heat generation curve for such a case can be calculated by adding the curves for the two separate models (Figs. 5 and 6) with appropriate weighting factors: $(1 - \varphi)$ for the continuous-production curve and φ for the single-event curve. For the latter case, the now-extant nuclides must be adjusted from 6.5 to 4.7 Æ before the present (implying a Δ varying somewhat with φ when the latter is large). Figure 7 shows the results of such calculations for various values of φ. If there were a period of inactivity preceding the terminal addition, the curves for small φ would be somewhat different, but for $\varphi \gtrsim 10^{-3}$ they would be essentially the same.

It can be seen that it is possible for matter so synthesized to have generated within itself as much as ~1000 cal g^{-1} of heat for times within a very few million years of the last addition, provided that that addition amounted to several per cent of the total matter. It is interesting that the nuclide Al^{26} [half-life 0.74 ± 0.03 My] (*43*) is almost solely responsible for the supplementary heat, as first suggested by Urey (*39*). [For Fe^{60}, the half-life used in the calculations, ~0.3 My (*44*), is now known to be too long by a factor of ~3 (*45*), making it much less important than indicated in the figures.]

It appears, then, that now-extinct radioactivity may have been important as a heat source in the early stages of the solar system under, and only under, "mixed" nucleosynthesis. With single-event nucleosynthesis, the bulk of the radiogenic heat would have been dissipated long before the condensation of the proto-bodies of the system. With simple continuous nucleosynthesis, the steady-state heat generation rate by nuclides with half-lives $\geqslant 10^5$ years would have been too small. Moreover, the "mixed" mechanism with a moderate value of φ (~10^{-2}–10^{-1}) makes it much easier to account for the radiogenic Xe^{129} in the chondrites.

Time Dependence of Nucleosynthesis

The arguments on radiogenic heat provide, as we have seen, one indication of "fine structure" in the time dependence of nucleosynthesis within the period of its course for the elements of the solar system. Not only is it likely that an individual supernova contributed substantially to "our" matter at the very end of the process, but it is probable that similar discrete major contributions occurred at other times as well. The

total number of important contributions may possibly have been quite small. Information about the time of the next-to-last contribution might conceivably be obtainable from more detailed studies of primary and extinct natural radionuclides, particularly if more representatives of the latter can be found.

The probable large magnitude of the last contribution suggests that our matter may contain only relatively small contributions from early nucleosynthetic events. This is suggested also by the independent argument that the early-synthesized elements introduced into the interstellar mixing-pot should have been removed steadily, both by incorporation into non-exploding stars (including the remnant cores of supernovae) and by reworking in other nucleosynthetically active stars. In the latter case, some of the "new" matter would be formed at the expense of "old" matter rather than of primordial hydrogen (and helium?).

To account for this factor, it is necessary to modify the differential equation (2) by the addition of a term representing the "death" of synthesized heavy nuclei by processes other than radioactive decay. As a first approximation (for the sake of mathematical tractability), we may consider that there is a constant probability $\lambda_\dagger$, the same for all nuclides, that a synthesized atom will be lost from the interstellar medium:

$$\frac{dN_i}{dt} = \rho_{i,0} e^{-\lambda_\star (t-f)} - \lambda_i N_i - \lambda_\dagger N_i \qquad (3)$$

It turns out that the resulting expressions for ratios such as $(N_5/N_8)_0$ and $(N_2/N_8)_0$ can be put into exactly the same form as those resulting from the solutions of equation (2), with $\lambda_\star - \lambda_\dagger$ substituted for $\lambda_\star$ at all points. Thus the resulting value of Δ will not be affected by the analysis described earlier. However, the value obtained for $\lambda_\star$ is really the value of $\lambda_\star - \lambda_\dagger$, so that $\lambda_\star$ must definitely be larger than the value obtained previously.

As usual, introduction of another parameter requires an additional piece or type of information for its evaluation. For this purpose, non-nuclear astrophysical evidence may be used to evaluate $\lambda_\star$ independently, and $\lambda_\dagger$ can then be obtained by difference. The time dependence of star formation has been the object of several studies (*46–50*). Salpeter (*49*) deduced that the rate of star formation is closely porportional to the density of interstellar gas, and proposed that the rate of formation of a heavy nuclide is proportional to the total mass of gas in the galaxy. For the latter he takes a result of J. H. Oort, based on radio-astronomy observations, that the present mass of gas in our galaxy is about 2% of the total mass of the galaxy. If the total time since the galaxy was all gas is $\Gamma = \mathrm{E} + \Delta + \Sigma \approx 14$ Æ, this would require a $\lambda_\star$ of $\sim$0.3 $Æ^{-1}$ to yield $e^{-\lambda_\star \Gamma} \approx 0.02$. If $\lambda_\star - \lambda_\dagger \approx 0.1$ $Æ^{-1}$, then $\lambda_\dagger$ would be about 0.2 $Æ^{-1}$. This means that a stable heavy atom would have a mean lifetime of about 5 Æ in interstellar space.

An even better approximation would be to assume that $\lambda_\dagger$, like the production rates ρ_i, varies proportionally with the rate of star formation:

$$\frac{dN_i}{dt} = \rho_{i,0}\, e^{-\lambda_\star (t-f)} - \lambda_i N_i - \lambda_{\dagger,0}\, e^{-\lambda_\star (t-f)}\, N_i \qquad (4)$$

Solutions to this equation would be considerably more complicated.

Other Cases of Extinct Natural Radioactivity

Although there are hundreds of nuclides which have been formed in natural processes but are now extinct, only those which have left effects which can now be detected in nature are designated as "extinct natural radionuclides" (*15*). The most definite type of such effect, a radiogenic excess of the daughter nuclide, is illustrated by the $I^{129} \rightarrow Xe^{129}$ transition. This case is favored by a moderately long half-life and an extremely high degree of chemical fractionation between the respective elements in the formation of the meteorites. If suffers the disadvantage, however, that the decay product could not accumulate until substantial cooling of the meteorite parent bodies had occurred. Thus for other nuclides with even somewhat shorter half-lives there is a possibility of observing extinct natural radioactivity. However, transitions such as Pu^{244} ($H = 7.6 \times 10^7$ y) $\rightarrow Th^{232}$, U^{236}($H = 2.39 \times 10^7$ y) $\rightarrow Th^{232}$, and Np^{237} ($H = 2.20 \times 10^6$ y) $\rightarrow Bi^{209}$ are unfavorable because the product does not have a stable or long-lived isotope, and those such as Sm^{146} ($H \approx 5 \times 10^7$ y) $\rightarrow Nd^{142}$ and Cm^{247}($H > 4 \times 10^7$ y and probably $> 9 \times 10^7$ y) $\rightarrow U^{235}$ because of the probable insufficiency of chemical fractionations between parent and daughter elements in meteorites.

Although a suggestion that a part of the Xe^{129} of the earth's atmosphere is radiogenic (*51*) has been regarded as questionable (*52*), "primordial" xenon is currently being found in some carbonaceous and enstatite chondrites (*18, 20–23*), which may lead to a determination of this point. According to the most plausible interpretation of the comparison between the xenon in the carbonaceous chondrite Murray and the xenon of the earth, about 5% of terrestrial Xe^{129} is due to I^{129} decay subsequent to isolation of the earth from cosmic xenon; and that event occurred about 50 M y after the isolation of the meteorites from cosmic xenon (*20*).

From the isotopic composition of primordial xenon, Kuroda (*53*) has argued the plausibility of earlier suggestions that some of the terrestrial Xe^{136} and other heavy Xe isotopes are a result of fission, most likely spontaneous, of heavy nuclei. The unusual mass-number distribution of the fissiogenic component suggests that now-extinct transuranium nuclides, especially Pu^{244} or Cm^{247} or both, are responsible. Goles and Anders (*54*) have concurred in this suggestion and have discussed the conclusions which might be drawn from xenon isotope studies relative to the chronology of the early solar system.

The moon's tenuous atmosphere may also contain—or may be made predominantly of—radiogenic Xe^{129} (*55*).

A search by Hess, *et al.* (*56*) for abnormality in the Ag^{107}/Ag^{109} ratio of meteoritic silver due to decay of now-extinct Pd^{107} ($H \sim 7 \times 10^6$ y) gave negative results. Very recently, however, Rama Murthy (*57*) has reported that the silver isolated from the troilite of an iron meteorite (Toluca) has an Ag^{107} content about 1% higher than terrestrial silver. The palladium-silver fractionation supposed to have taken place while Pd^{107} was still present was that in the formation of the metallic phase rather than that in the later segregation of the troilite nodules. Making certain assumptions, it was calculated that this fractionation would have taken

place 84 My after a single-event nucleosynthesis or only 10 My after simple continuous synthesis. However, it was pointed out that the abundance of Pd^{107} at the end of long-term nucleosynthesis would depend to a large extent on its production in the last contributing supernova. Assuming the terminal Pd^{107} abundance to be 10% of its single-event value, the interval between the terminal contribution and the fractionation was calculated to be 45 M y. This is unexpectedly short, but gratifyingly consistent with the hypothesis of "mixed" nucleosynthesis with $\varphi \sim 0.1$, with intense short-lived heating mainly by Al^{26}, and with the longer cooling periods before the chondrites began to retain Xe^{129}.

A search for enhancement of the Tl^{205}/Tl^{203} ratio as a result of Pb^{205} decay ($H \approx 2.4 \times 10^7$ y) has recently been reported by Anders and Stevens (*58*), who examined the thallium from two typical chondrites (including Richardton), an enstatite chondrite, a carbonaceous chondrite, and the troilite of two siderites (Toluca and Canyon Diablo). In all cases the isotopic composition was normal within experimental uncertainty (about 1% of the ratio). From these "negative" results, of which that on Canyon Diablo troilite was most significant, it was calculated using certain assumptions that the decay interval was ≥ 0.10 Æ on the continuous nucleosynthesis hypothesis and ≥ 0.30 Æ on that of a single nucleosynthetic event. The "mixed" nucleosynthesis model would, of course, give an intermediate limit. El-Badry and Kohman (*59*) have pointed out that the large variations in the thallium content of meteoritic troilite make it quite likely that samples with more favorable (higher) Pb/Tl ratios than those so far examined may well exist, so that the incorporation of detectable levels of radiogenic Tl^{205} may well have occurred.

Reed, *et al.* (*60*) point out that the absence of a detectable amount of Bi^{209} in an achondrite (Nuevo Laredo) implies that there was essentially no Np^{237} present at the time of a fractionation which removed bismuth from the material of the meteorite. Assuming single-event nucleosynthesis, they calculate a decay interval of $\geq$ 16 M y preceding that fractionation. The limit would be somewhat lower on the basis of "mixed" nucleosynthesis, and no limit would be implied on the simple continuous model.

Terrestrial minerals display a greater variety and degree of chemical fractionation than do the meteorites, and might therefore offer additional possibilities for the detection of extinct natural radioactivity (*15*). However, the fractionations leading to accessible rocks and minerals would have occurred considerably later, and hence only quite long-lived nuclides would come into question. Very recently there has been reported by Cherdyntsev and collaborators (*61*) an observation of a 50% excess of U^{235} in the uranium from a terrestrial magnetite, suggesting the presence at its formation of some transuranium nuclide of the $4n + 3$ series, most likely Cm^{247}. If this is indeed the explanation, the half-life of Cm^{247} must be at least $\sim 10^8$ years, and it may be just barely or even not quite extinct. It has been pointed out (*62*) that such a situation would provide an important method for dating very early geologic events. However, a search for U^{235} enhancement in two rare-earth minerals ~1 Æ old produced negative results (*63*).

Very few blank spots in the chart of nuclides remain which might be filled by nuclides with half-lives $\gtrsim 10^7$ years. One possibility is Mn^{53}, which has been observed in meteorites as a result of cosmic-ray production (*64*), and whose half-life is estimated theoretically as $\sim 10^7$ y (*65*). The half-life of Hf^{182} has just been determined as 8.5×10^6 y (*66*). Since hafnium is lithophile and tungsten is siderophile, W^{182} might be enhanced in stone meteorites. Fissiogenic krypton isotopes from extinct Pu^{244} and Cm^{247} might be detectable in meteorites and in the earth's atmosphere. The apparently short time between the end of large-scale nuclear reactions and the beginning of large-scale chemical reactions among the elements of the solar system indicates that all nuclides with half-lives greater than $\sim 10^6$ years should be regarded as potentially extinct natural radionuclides.

The year 1960 has seen the promotion of "extinct natural radioactivity" from a speculative concept to an active and important field of research in cosmochemistry.

Acknowledgments

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Discussions with G. G. Goles and R. A. Fish led to the modifications [(3) and (4)] of the differential equation (2) for galactic nucleosynthesis.

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Redetermination of the Half-Life of Carbon-14

A more accurate value for the half-life of carbon-14, important in geological and archeological dating, has recently been obtained by the National Bureau of Standards. The new value is 5760 years, as compared with the previously accepted value of 5568 years.

In performing the redetermination, high-specific-activity carbon dioxide was quantitatively diluted for counting in length-compensated internal gas counters in the Geiger and proportional regions. Mass spectrometric analyses of parts of the undiluted gas sample were made to determine the isotopic abundance of carbon-14, which was found to be approximately 44 atom-percent. Intercomparative measurements of the isotopic abundance were also carried out with the United Kingdom Atomic Energy Authority.

The redetermined value is in fairly good agreement with the value of 5900 ± 250 years obtained at the Bureau in 1953. The value of 5568 years that has been in use was somewhat arbitrarily chosen. Because of the wide range in measured values of the half-life of carbon 14—from 4700 to 7200 years—a weighted average of three values determined by gas counting and by mass spectrometric analysis was tentatively accepted for the purpose of radiocarbon dating of archeological samples. The present measurements have led to the conclusion that the uncertainties in the values obtained experimentally may have arisen almost entirely from adsorption effects. The change, however, is less than the previously stated experimental error.

National Bureau of Standards Office of Technical Information

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The Karlsruhe Congress: A Centennial Retrospect

On September 3, 1860, there gathered in the resort city of Karlsruhe about 140 European chemists, assembled to seek a meeting of minds regarding some of the problems responsible for the chaotic state of mid-nineteenth century chemistry. Friedrich August Kekulé, then a professor at the University of Ghent and one of the promising figures among the younger chemists, was the leading stimulus in bringing about the conference. He had gained the backing, a year earlier, of Adolf Wurtz and Carl Weltzein. The latter, professor at the Technische Hochschule in Karlsruhe, shouldered the burden connected with arrangements for the meeting (*1*). Forty-five prominent chemists[1] permitted their names to be used on the invitations sent out on July 10, 1860.

The Chaos That was Chemistry

The meaning of such terms as "atom," "molecule," and "equivalent" had been in a state of chaos during the previous 50 years. Failure to agree on a uniform system of atomic weights meant that formulas varied from laboratory to laboratory. As an example, the atomic weight of 8 was used by some chemists for oxygen, 16 by others. Atomic weights of 6 and 12 were both in use for carbon. Consequently, formulas for the same substance could not be in agreement. Even when there was agreement on the empirical formula there was still a variation in ideas on the nature of combination in compounds. When Kekulé prepared the first volume of his textbook (*2*) in 1861 he listed 19 different formulas for acetic acid, all of them to be found in the literature (Fig. 1). In an era such as this, chemistry was not only complicated but without sound foundations.

The whole problem began a half century earlier with the introduction of Dalton's atomic theory. Dalton, in his early work, had introduced a list of atomic weights which proved highly inaccurate and quite inadequate. In 1808 Gay-Lussac (*3*), in his paper on combining volumes, published information which suggested that two atoms of hydrogen combine with one of oxygen. This led to use of the formula H_2O by Berzelius although Dalton preferred the formula HO. While the law of combining volumes actually bolstered Dalton's atomic theory, Dalton refused to consider it of significance and questioned the reliability of Gay-Lussac's work.

The Italian physicist Avogadro (*4*) brought about a reconciliation of Gay-Lussac's law and Dalton's atomic theory in 1811 when he suggested that equal volumes of gases at the same temperature and pressure contain the same number of molecules. He then went on to show that the apparent discrepancies between the law of combining volumes and the atomic theory might be reconciled if elemental gas molecules were considered to be polyatomic. Nowhere did Avogadro argue that all elemental gases were diatomic, but since each of

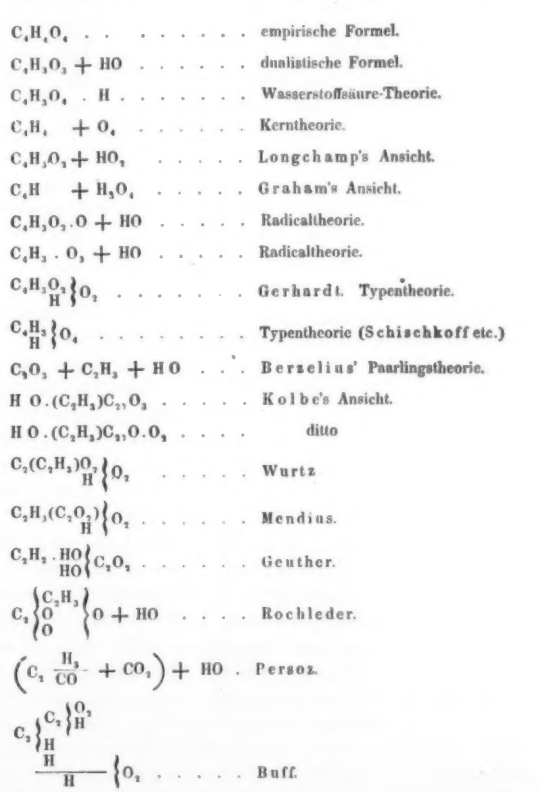
$C_4H_4O_4$ . . . . . empirische Formel.
$C_4H_3O_3 + HO$ . . . . . dualistische Formel.
$C_4H_3O_4 \cdot H$ . . . . . Wasserstoffsäure-Theorie.
$C_4H_4 + O_4$ . . . . . Kerntheorie.
$C_4H_3O_3 + HO_2$ . . . . . Longchamp's Ansicht.
$C_4H + H_3O_4$ . . . . . Graham's Ansicht.
$C_4H_3O_2 \cdot O + HO$ . . . . . Radicaltheorie.
$C_4H_3 \cdot O_3 + HO$ . . . . . Radicaltheorie.
$C_4H_3O_2\}O_2$ . . . . . Gerhardt. Typentheorie.
$C_4H_3\}O_4$ . . . . . Typentheorie (Schischkoff etc.)
$C_3O_3 + C_2H_3 + HO$ . . . Berzelius' Paarlingstheorie.
$HO.(C_2H_3)C_2O_3$ . . . . . Kolbe's Ansicht.
$HO.(C_2H_3)C_2O.O_3$ . . . . ditto
$C_2(C_2H_3)O_2\}O_2$ . . . . . Wurtz
$C_2H_3(C_2O_2)\}O_2$ . . . . . Mendius.
$C_2H_2 \cdot HO\}C_2O_2$ . . . . . Geuther.
$C_2\{C_2H_3\}O + HO$ . . . . Rochleder.
$\left(C_2\frac{H_2}{CO} + CO_2\right) + HO$. Persoz.
$C_2\{C_2\}O_2\}H$ Buff. . . . . Buff.

Figure 1. Tabulation of formulae for acetic acid in use before 1861. From F. A. Kekule, "Lehrbuch der organischen Chemie," 1861.

Presented before the Division of History of Chemistry at the 138th Meeting of the American Chemical Society, New York City, September 13, 1960.

[1] The following names were appended to the invitations sent out by Weltzein (asterisks show those who did attend the meeting). Babo,* Balard, Beketoff, Boussingault,* Brodie, Bunsen,* Bussy, Cahours, Cannizzaro,* H. Deville, Dumas,* Engelhardt, O. L. Erdmann,* Fehling,* Frankland, Fremy, Fritzsche, Hlasiwetz,* Hofmann, Kekulé,* Kopp,* Liebig, Malaguti, Marignac,* Mitscherlich, Odling,* Pasteur, Payen, Pebal,* Peligot, Pelouse, Piria, Regnault, Roscoe,* Schötter, Socoloff, Staedeler, Stas,* Strecker,* Weltzein,* Will,* Williamson, Wöhler, Wurtz,* Zinin.*

his examples was a diatomic molecule, the few chemists who attempted to deal with the hypothesis supposed that molecules of all elemental gases or vapors were diatomic.

Nearly all chemists, however, found Avogadro incomprehensible or unconvincing; the hypothesis, together with Ampère's independent reiteration of it in 1814, lay in the discard for a half century. Berzelius, leading student of electrochemistry, was of the opinion that atoms were held together incompounds by electrical forces; hence, two atoms of the same element would repel each other and diatomic molecules would not be possible. Although Berzelius utilized the principle that differential elemental gases contained the same number of atoms per volume, he could not accept the notion that compound gases also contained the same number of molecules.

When Dumas (*5*) developed his method for vapor density determinations in 1826, he found disagreement between atomic weights calculated from vapor densities and the atomic weights for the same elements reported from purely chemical investigations. Although Gaudin (*6*) suggested that elemental gas molecules might contain other than two atoms, Dumas and everyone else remained unconvinced and vapor densities played no further part in the atomic weight problem.

The next 30 years saw the dramatic rise of organic chemistry, with an attendant intensification of the problem of inconsistent formulas. The various radical and type theories of the period confused rather than clarified the problem.

Despite the excellent analytical work of Berzelius, and his shrewd deductions, there was a hesitancy to accept atomic weights as having value in the calculation of formulas. Following Leopold Gmelin, many chemists felt more comfortable with equivalent weights and used them in dealing with proportionate relationships in chemical compounds. Gerhardt, early in the forties, reiterated the greater significance of atomic over equivalent weights, but recognized certain inconsistencies in the Berzelius table and made corrections. However, in correctly halving the Berzelius' values for silver, sodium, and potassium, he also halved the correct values for zinc, calcium, and other divalent metals.

The work of Wurtz and Hofmann on amines led to the recognition around 1850 of the ammonia type of compound. About the same time, Williamson's work led to recognition of the water type of compound in the alcohols and ethers. Gerhardt added the hydrogen and hydrogen chloride types shortly thereafter and sought to develop a new type theory for the classification of organic compounds.

Kolbe's electrochemical studies on fatty acids led him to grope toward a primitive sort of structural formulation. Frankland, as a consequence of his work with zinc alkyls, was arriving at the valence concept for organic groups and metallic atoms. Kekulé and Couper independently extended the valence concept when they recognized the quadrivalency of the carbon atom. Both further recognized that carbon atoms might satisfy a part of their valency by carbon-to-carbon bonding. Kekulé and Couper each sought to deal with structure in organic compounds on the basis of valence, Couper's structures bearing a close resemblance to those which later came into common use.

It becomes readily apparent that the discipline of chemistry was in a state of extreme chaos at mid-century and was, in fact, ripe for reform. The older chemists like Berzelius, Liebig, Dumas, Gmelin, and Mitscherlich were giving ground to a new generation of chemists. Some, particularly Laurent and Gerhardt, were openly contemptuous of authority. Others like Kekulé were respectful of the past, but not prevented by it from examining unique new concepts. It was in this atmosphere that the Karlsruhe Congress was called.

Following the customary preliminaries, the Congress settled down to business with the appointment of a steering committee, with Kopp as chairman, Béchamp, Canizzaro (Fig. 2), Erdmann, Fresenius, Kekulé, Schischkoff, Strecker, and Wurtz, to draft a set of questions which would form the basis for the second day's discussion. This session was spent in an examination of the various ways in which the terms "atom," "molecule," "radical," and "equivalent" were used. There was no unanimity of opinion and no decisions were reached.

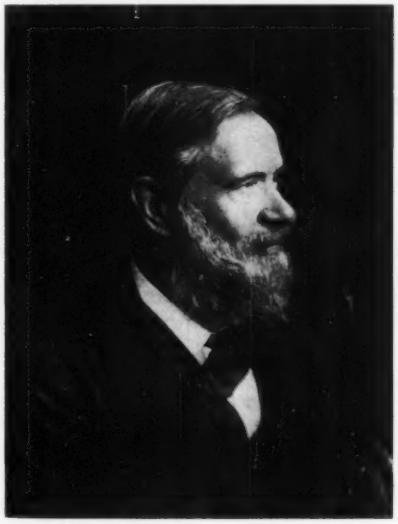

Figure 2. Stanislao Cannizzaro.

Cannizzaro's Influence

At the last meeting on September 5 the steering committee proposed three questions for discussion.[2] One of them raised the advisability of returning to the principles of Berzelius. Cannizzaro spoke against the proposal, pointing out that Gerhardt had set chemistry on the right track by basing molecular weights on the concept introduced by Avogadro and Ampère. Hence, it was important to utilize Gerhardt's contributions rather than retreat to the position of Berzelius.

Cannizzaro, then at the University of Genoa, had published two years earlier a sketch of his method of teaching chemistry (*7*). In this paper he revealed

[2] For details see C. DE MILT, *Chymia*, **1**, 165 (1948), or better, KARL ENGLER in the "Festgabe zum Jubililäum der vierzigjährigen Regierung Seiner Königlichen Hoheit des Grossherzogs Friedrich von Baden," Karlsruhe, 1892, pp. 346–55. The latter paper carries the official report of the meeting which was prepared by Wurtz and filed in the Archives of the Technical School in Karlsruhe.

how, by an examination of historical theories and experiments, it was possible "to lead my students to the conviction which I have reached myself" (*8*). He described the significance of certain contributions of Gay-Lussac, Avogadro, Ampère, Berzelius, Dumas, Regnault, Gerhardt, and Laurent, and showed in his lectures, how these contributions were useful in the clarification of the subject matter.

Cannizzaro's speech before the Congress emphasized the significance of Gerhardt's work, in which the latter based his molecular weights on the hypothesis of Avogadro and Ampère. Attention was called to the work of Dumas on vapor densities and the reasoning of Gaudin on the size of molecules of elemental gases. The application of Avogadro's hypothesis to the determination of atomic weights was explained and a plea was made for the adoption of atomic and molecular weights based on the application of these principles.

During the discussion which followed Strecker announced his intention of adopting the proposed atomic weights and Kekulé agreed, but with certain reservations. Kopp and Erdmann argued, however, that votes must not be taken on scientific questions and the meeting closed on a note of uncertainty. At that time Angelo Pavesi, professor of chemistry at Pavia and follower of Cannizzaro, distributed reprints of Cannizzaro's paper of 1858. While many copies were doubtless consigned to the trash can, a few were read with understanding. Lothar Meyer was profoundly influenced (*9*) and utilized Cannizzaro's suggestions as the basis for his own textbook, "Die Modernen Theorien der Chemie" (1864). Mendeleev was another who made extensive use of the proposals as had Newlands before him.

It is impossible to overemphasize the role of Cannizzaro in bringing about the acceptance of Avogadro's hypothesis, and thereby pointing chemistry along a fruitful path. By comparing the density of gases and vapors with the density of hydrogen, and by accepting the diatomic character of the hydrogen molecule, he was able to show how correct molecular weights might be determined. By accepting the comparative densities observed by use of the balance, and by refusing to entertain preconceived notions regarding molecular composition, he was able to avoid the uncertainties which plagued Dumas with respect to the vapors of sulfur, mercury, phosphorus, and arsenic. It became increasingly obvious that molecular size might be dealt with with certainty.

As a consequence of the certainty of molecular weights, it now became possible to distinguish between empirical and molecular formulas. Hydrocarbons, alcohols, organic acids, aromatic compounds, and virtually all of the simpler organic molecules could be formulated correctly.

Structural Chemistry Possible

With the advent of reliable molecular formulas it was possible to make the step to structural chemistry. Although Kekulé and Couper had grappled with the problem as early as 1858, it could not be convincingly handled until molecular formulas were no longer open to question. During the eighteen sixties a vigorous period of chemical architecture developed through the work of such chemists as Kekulé and Butlerov. This type of thought paved the way for the successful solution of the molecular basis for optical activity which had plagued chemists since the forties. Van't Hoff and Le Bel successfully dealt with the problem of the asymmetric carbon atom in 1874. Their work was successfully extended during the next decades, particularly in the hands of J. Wislicenus and, in the case of the sugars, of Emil Fischer.

The last four decades of the 19th century were extremely fruitful ones for the development of organic chemistry. This is evidenced by the successful development of the synthetic dye industry, the branching out into the field of synthetic drugs, and the development of a significant indicator theory before the end of the century. The successful elucidation of the structure of such natural dyes as alizarin and indigo, and of the sugars, purines, and terpenes, reveals the power held by the organic chemists of the day.

It is true that the successes in structural chemistry did not extend into the inorganic field until the end of the century. The attempt of Blomstrand and of Jørgensen to deal with structure of inorganic compounds was a miserable failure. There was an inability to realize that whereas organic structures involve a very limited number of elements where carbon atoms serve as a central skeleton, inorganic compounds are made up of a greater variety of elements with no single element serving as a central core. The problem only found resolution in the 1890's when Alfred Werner arrived at the concept of the coordination compound and began dealing with coordination complexes from the standpoint of geometric structure. However, it was necessary for Werner to introduce the concept of primary and secondary valences. Since a sound theory of chemical bonding only began to develop after the rise of the structural atom around 1914, it was difficult for inorganic chemists to reconcile themselves to the demands of primary and secondary valences.

Prototype of International Meetings

Besides its impact on the development of chemical theory, the Karlsruhe Congress represents the first international chemical meeting. Although it failed to mark the introduction of a regular series of international meetings it represented a preliminary step among chemists to seek, through a large gathering representative of many countries, solutions to problems which are difficult to resolve on the local level. As chemistry advanced during subsequent decades more of these problems began to be apparent, leading to new international meetings.

By 1889 the problem of naming organic compounds was sufficiently acute to demand the attention of the International Congress of Chemists which assembled in Paris in that year. A commission was set up at that time to make an interim study. A report was submitted at the Geneva meeting of the Congress three years later. This was approved by about forty chemists in attendance, forming the basis of the Geneva system of organic nomenclature.

The International Congress of Chemists continued to meet at regular intervals until World War I. Following the termination of the war, the need for an international organization of chemists was recognized

in the formation of the International Union of Pure and Applied Chemistry at meetings of an international group held in London and Brussels during July in 1919. This organization has continued active in dealing with nomenclature and other problems of world-wide interest. International Congresses and Conferences have been held, except during World War II, at intervals of several years.

The Karlsruhe Congress, in retrospect, had significant consequences, even though it appeared to end with little accomplished (*10*). There is no doubt that the problems facing the conferees would have been solved without a Karlsruhe Congress, but they would have given ground more slowly. The Congress dramatized the importance of Avogadro's hypothesis in the minds of a few of the leading younger chemists, resulting in the application of a sound principle which would make possible the dramatic progress in chemistry during the next four decades of the century.

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The Mole and Related Quantities

The scale of "atomic weights" based on O = 16 was introduced under the influence of Berzelius early in the nineteenth century and has been used ever since by chemists. In 1929 Giauque and Johnston (*1*) discovered the isotopes ^{18}O and ^{17}O of the hitherto only known oxygen nuclide ^{16}O. From that time physicists have for obvious reasons used the scale $^{16}O = 16$. The chemical scale and the physical scale differ by the factor 1.000275. The difference between the two scales has been a source of increasing inconvenience and indeed irritation to both chemists and physicists. For nearly four years there have been discussions (*2*) on the possibility of finding a scale acceptable to both, discussions which at last have resulted in success. The physicists for technical reasons which need not be elaborated required a scale based on a pure nuclide with mass number a multiple of four. The chemists on the other hand, in order not to have to scrap the extensive tables of molar quantities, were prepared to accept a change only if it was less than 1 part in 10^4. These conditions ruled out both existing scales. In 1957 Olander and Nier independently proposed (*2*) the scale $^{12}C = 12$ and this has now been accepted by both chemists and physicists. The International Union of Chemistry at its meeting of 1959 in Munich decided (*3*) that it would formally introduce the scale $^{12}C = 12$ at its next biennial meeting of 1961 in Montreal provided that meanwhile the scale was formally endorsed by the International Union of Physics. This was done (*4*) at the latter Union's meeting in September, 1960, in Ottawa. A comparison between the two obsolescent scales and the new unified scale is summarized in the following table:

	Old physical scale	Old chemical scale	New unified scale
^{16}O	16 exactly	15.99560	15.99491
O	.....	16 exactly	15.9999
^{12}C	12.00382	12.00052	12 exactly
C	.....	12.011	12.010
Ag	.....	107.880[a]	107.875
		107.873[b]	107.868

[a] Value given in the 1959 International Table of Atomic Weights.
[b] Value obtained from the recent coulometric determination at the National Bureau of Standards (*7*) of the electrochemical equivalent of silver.

It will be observed that the change for chemists is only 43 parts per million and this is numerically trivial, but this change is a tremendous triumph of reasonableness over confusion. The change also provides an occasion for tidying up nomenclature related to the scale. In particular it is hoped that the term "atomic weight"

may fall into disuse so that eventually it may become unnecessary to explain to every novice that "atomic weight" does not mean weight of an atom.

The following text is an outline of recommendations on terminology and notation already endorsed (*4*) by the International Union of Physics. Some of these have also been endorsed by the International Union of Chemistry; it seems probable that this body will make similar if not identical recommendations. Most of these recommendations have also been endorsed by the International Standards Organization's Technical Committee on Quantities, Units, Symbols, Conversion Factors, and Conversion Tables.

Mole: Chemists' "Amount of Substance"

We have all been taught at an early age that mass and weight are different quantities although at a given place their ratio is constant. During the past score of years the view has been accepted by a rapidly increasing number of physicists and chemists that there is a third quantity different from mass and weight but proportional to both. This quantity was first named (*5*) "Stoffmenge" in German and the English translation is "amount of substance." Admittedly the amount of a pure dry solid substance is usually measured by weighing, but there are numerous other ways of measuring amount of substance. The amount of a gas (or beer!) is often determined by a measurement of volume. The amount of a substance in solution is often determined by titration or by colorimetry or by polarography. The amount of a radioactive substance may be determined by a Geiger counter. The amount of a drug may be determined by its physiological action. This list is not exhaustive. Weight is a measure of gravitational pull and mass determines the redistribution of momentum in a collision. It is clear that amount of substance determinable by any of the mentioned techniques is no more identical with mass or with weight than these two quantities are identical with each other. The unit of amount of substance most used and among chemists almost universally used is the mole. We continue with a list of definitions related to the mole and to the new unified scale.

A *nuclide* is an atomic species defined by both atomic number and mass. Two or more nuclides having the same atomic number but different atomic masses are called isotopes. (A single isotope is, like a single twin, a contradiction in terms.)

The mass of an atom of a nuclide, or the average mass of an atom in an element, is for convenience expressed as the ratio of this mass to one-twelfth of the mass of a neutral atom of ^{12}C. This ratio is called the *relative nuclidic mass* of the nuclide or the *relative atomic mass* of the element. The mass equal to one-twelfth that of a neutral atom of ^{12}C is denoted by m_u in analogy with m_p for the mass of a proton, m_n for the mass of a neutron, and m_e for the mass of an electron. Alternatively this same quantity may be regarded as a unit of mass for the new unified scale and it is then denoted by u. We may thus write either for the relative atomic mass of ordinary silver

$$m_{Ag}/m_u = 107.87$$

or for the mass of an atom of ordinary silver

$$m_{Ag} = 107.87\text{u}$$

where

$$m_u = 1\text{u} = 1.66037 \times 10^{-24}\ \text{g}$$

The mole is the amount of substance containing the same number of molecules (or atoms or radicals or ions or electrons as the case may be) as there are atoms in 12 grams of ^{12}C.

Avogadro's constant denoted by L or N_A describes the number of molecules per amount of substance. Its value on the unified scale is

$$L = 6.02278 \times 10^{23}\ \text{mole}^{-1}$$
$$L^{-1} = 1.66037 \times 10^{-24}\ \text{mole}$$

Faraday's constant, denoted by F, describes the charge of a mole of electrons and is equal to the product of Avogadro's constant L and the elementary charge e. On the unified scale

$$F = Le = (6.02278 \times 10^{23}\ \text{mole}^{-1})\ (1.60209 \times 10^{-19})\ \text{coulombs}$$
$$= 96490\ \text{coulombs-mole}^{-1}$$

The *gas constant* denoted by R is equal to the product of Avogadro's constant L and Boltzmann's constant k. On the unified scale and on the temperature scale prescribed by the General Conference on Weights and Measures with the triple point of water defined (*6*) as $273.16°K$:

$$R = Lk = (6.02278 \times 10^{23}\ \text{mole}^{-1})\ (1.38049 \times 10^{-16})\ \text{erg}\ °\text{K}^{-1}$$
$$= 8.3144 \times 10^{7}\ \text{erg}\ °\text{K}^{-1}\ \text{mole}^{-1}$$
$$= 8.3144\ \text{joules}\ °\text{K}^{-1}\ \text{mole}^{-1}$$

Terminology Still Needed

There remains at least one unresolved question of terminology. There is a need for a word to denote per amount of substance analogous to *specific* meaning per mass and *density* meaning per volume. This need has been stressed especially by French-speaking scientists but no proposal has yet been made to fill the gap. The present author with some diffidence suggests the word *proper* (French "propre," German "eigen") because he knows of no better word. If this suggestion were adopted we should say that the "proper energy" of a substance was so many joules per mole rather than saying that the "molar energy" is so many joules per mole, which is as clumsy as if we spoke of "gram energy" instead of "specific energy."

I am grateful to Dr. Stille for expert advice concerning the new values of the general constants.

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A thermoanalytical kinetic experiment

The Catalyzed Reaction of Phenyl Isocyanate with Butanol

The rate at which heat is absorbed or evolved during a chemical process can give a rapid and convenient estimate of reaction kinetics. Various calorimetric methods have been described in the literature (*1*), demanding, for the most part, elaborate and expensive apparatus and a complicated interpretation of thermal data. Recent methods applied most successfully and precisely toward this end have been differential enthalpic analysis (DEA) and differential thermal analysis (DTA). Both these techniques employ a differential thermocouple immersed in reactive and inert materials of approximately equivalent heat capacity, and temperature differences during reaction are sensed by a recording device. In DEA, described by Eyraud (*2*), temperature differences between the active and reference substances are maintained at zero by supplying heat to either during a reaction. The amount of heat input is recorded. In DTA (*3*) the temperature difference between materials is recorded directly as the environmental temperature is changing. A smooth closed curve results, the area beneath proportional to the heat of reaction. This latter technique has proved of greater value in the consideration of reaction kinetics. Thus, Kissinger has developed the relationship between the "shape index" of a differential thermogram and the order of reaction in solid state reactions (*4*), and Borchardt and Daniels have derived rate constants and activation energies from single curves through the application of DTA to reactions in solution (*5*). These methods are, however, limited to specific types of reactions not often encountered in the common organic laboratory; and the equipment required, though relatively simple in the Borchardt and Daniels experiment, may not be immediately available or indeed warranted by the nature of the experiment.

Thermal analysis (TA) is the natural precursor of the forementioned methods. It relies solely on the rate and extent of development of an exotherm upon rapid mixing of reactants and hence requires only the most rudimentary apparatus. The equipment used in the present experiment is that found in any conventional laboratory: a small Dewar flask, a thermometer calibrated in 0.1°C units, a magnetic stirrer, and a stop-watch.

The Theory of Thermal Analysis

The observed rate of temperature change in an exothermic reaction, under approximately adiabatic conditions, may be related to the rate of change of reactant concentration by the expression,

$$\frac{dT}{dt} = -\frac{\Delta H}{c}\frac{dn}{dt} \tag{1}$$

where ΔH is the heat of reaction and c is the calorimeter constant. The initial rate of temperature change is similarly related to the initial concentration change:

$$\left(\frac{dT}{dt}\right)_0 = -\frac{\Delta H}{c}\left(\frac{dn}{dt}\right)_0 \tag{2}$$

Integrating (1):

$$\Delta T = -\Delta H/c \int_0^{\infty} (dn/dt)dt = -\Delta H/c(n) \tag{3}$$

and dividing (2) by (3):

$$\frac{(dT/dt)_0}{\Delta T} = \frac{-\Delta H/c(dn/dt)_0}{-\Delta H/c\,(n)}$$

or,

$$(dn/dt)_0 = (dT/dt)_0 \cdot (n)/\Delta T \tag{4}$$

This states that the initial reaction rate is directly proportional to the initial temperature rate and inversely proportional to the total temperature rise. The reaction rate is also proportional to the rate constant, thus (*6*):

$$(dn/dt)_0 = k(n)^x \tag{5}$$

where k is the rate constant at the temperature of reaction initiation, x is the order of reaction, and n is the initial concentration of reactant. Substituting in equation (4):

$$k = \left(\frac{dT}{dt}\right)_0 \frac{(n)^{1-x}}{\Delta T} \tag{6}$$

and for a first order reaction,

$$k = \left(\frac{dT}{dt}\right)_0 \frac{1}{\Delta T} \tag{7}$$

Thus, for a first order (or "pseudo-unimolecular") reaction, the initial slope of a temperature-time curve divided by the total temperature rise is simply equal to the rate constant for reaction. This assumes that the heat of solution (or dilution) of the last introduced reactant is negligible compared to the heat of reaction. A typical temperature-time curve is illustrated in Figure 1. To determine the initial slope, a smooth curve is fitted to the raw "stop-watch" data and graphically differentiated, extrapolating to the zero intercept. It may be expedient, however, to feed raw data into a programmed computer, thus obviating a great part of human error.

Contribution No. 196 from the Research Center of the United States Rubber Company, Wayne, New Jersey.

The Experiment: Iron (III) Acetylacetonate Catalysis in the Reaction of Phenyl Isocyanate and *n*-Butanol

Aromatic isocyanates react rapidly with alcohols to form, predominantly, urethanes:

$$\text{Ar—NCO} + \text{R—OH} \rightarrow \text{Ar—NHCOO—R}$$

The reaction of diisocyanates with diols and polyols to give polyurethanes has created an entire new industry (elastomers, foams, coatings, etc.). Fortunately, the reaction above is not simple, but many side reactions do occur to varying extents, creating materials of wide property range. Therefore, catalysts are employed to promote certain reactions in polymerizing media and hence minimize others. Iron (III) acetylacetonate is one such catalyst, specific to the urethane reaction (*7*), and has found wide use. However, catalytic parameters have not been described and, for that matter, are rare for almost all known urethane catalysts. In a mechanistic discussion at these laboratories, it was postulated that the iron complex might accommodate more than one isocyanate group in a transition state intermediate, and this study was undertaken with that feature in mind. We also desired to set up a method for the determination of catalytic parameters for urethane catalysts in general.

Iron (III) acetylacetonate was prepared by mixing an ammoniated solution of 1,3-pentanedione with an equivalent amount (one-third molar ratio) of iron (III) sulfate hydrate. The red precipitate was filtered and recrystallized twice from benzene and petroleum ether, mp 187–188°. Quantities of the catalyst were dissolved in dried, distilled *n*-butanol.

Exactly 100.0 ml of the butanol-catalyst solution were placed in a small Dewar flask fitted with a magnetic stirrer, vented stopper, thermometer calibrated in 0.1° units, and a small calibrated rapid delivery addition funnel. A hypodermic syringe may be used in place of the latter. The initial temperature of the solution in the Dewar was adjusted to 24.5° with the aid of a cold finger and, at this temperature, between 1.0 and 3.0 ml of freshly distilled phenyl isocyanate were added rapidly. Temperature was recorded every 15 seconds thereafter up to four minutes, then leveling temperatures were recorded at one minute intervals up to 10 minutes.

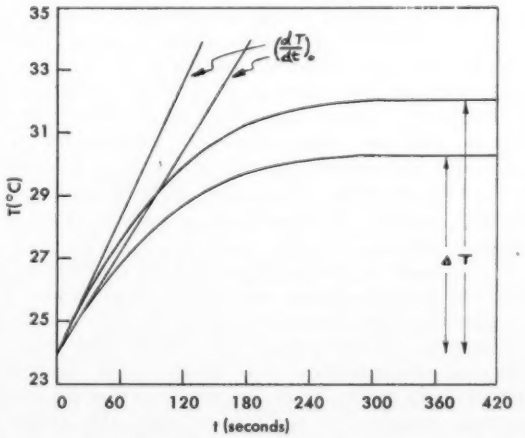

Figure 1. Typical temperature-time curves for the reaction or phenyl isocyanate with *n*-butanol.

All reactions investigated were essentially complete well within that 10-minute period. Typical smoothed temperature-time curves are shown in Figure 1. The data were analyzed via a Burroughs E-101 computer. In this manner, as many as 30 experiments were run (and 30 rate constants calculated) in a single day.

Catalytic coefficients may be calculated from the Brønsted relationship (*8*):

$$k = k_0 + k_c[\text{Cat}]^x \tag{8}$$

where k is the rate constant for reaction, k_0 the uncatalyzed rate constant, k_c the catalyst coefficient, [Cat] the catalyst concentration, and x the order of catalysis. In logarithmic form:

$$\log (k - k_0) = \log k_c + x \log [\text{Cat}] \tag{9}$$

Figure 2 shows the results, plotted as indicated by equation (9). The slope is the order of catalysis and the intercept the logarithm of the catalyst coefficient. The catalytic equation

$$-\log (k - k_0) = 0.0305 - 0.7739 \log [\text{Cat}]$$

$$R = 0.933$$

$$S_e = 0.072$$

where R is the correlation coefficient and S_e the standard error, shows a slope term close enough to unity to rule out the possibility of multiple occlusion of isocyanate in the transition state. The catalytic coefficient, $k_c = 0.932$ l/mole-sec, is useful for future comparison of other catalysts.

This method is, of course, subject to low precision of measurement which can be minimized by the rapid accumulation of replicate data and statistical analysis. In addition, it is best suited for reactions with half lives of no less than 30 seconds and no more than three minutes, to obviate, respectively, temperature measurement lag and heat losses. ΔT should be less than 10°C, the average temperature lying fairly close to ambient for this latter reason. As mentioned before, heats of solution and dilution should be negligible compared to heats of reaction.

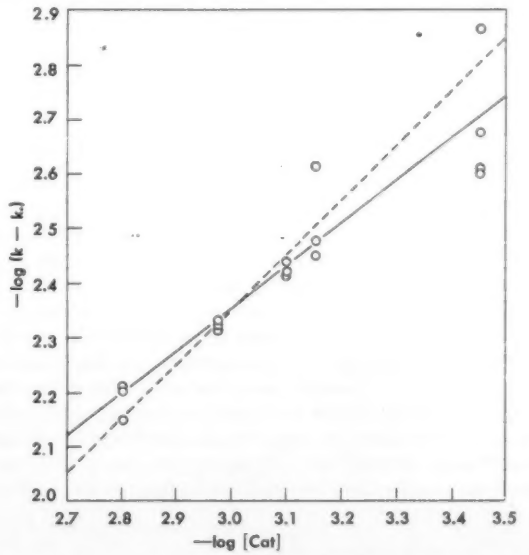

Figure 2. Bronsted plot for $Fe(CH_3COCHCOCH_3)_3$ catalysis of phyenl isocyanate and *n*-butanol; least squares slope ——, unit slope, - - - - -.

Acknowledgment

The author wishes to thank Dr. Hans Borchardt of the General Electric Company for his helpful discussions.

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Non-Stoichiometry in Non-Metallic Compounds

The laws of definite combining proportions and simple combining ratios as applied to condensed phases are sometimes taken too rigorously. For the gaseous molecule NO, the smallest change that one can produce in this molecule is either the addition or removal of one atom. For example, one could add one nitrogen atom to produce N_2O or one oxygen atom to produce NO_2, both of which are compounds with properties drastically different from those of nitric oxide. As the atom is the smallest unit which can be added or removed from a molecule, it is clear that the smallest change that one can produce in a gaseous molecule will produce a large change in its properties. If, on the other hand, we consider a molecule of solid sodium chloride, for example a single crystal weighing 10 g, its formula would be $Na_{10^{23}}Cl_{10^{23}}$. Here again, the smallest change that one can bring about is the addition or removal of one atom. If we remove one chlorine atom, the new formula is now $Na_{10^{23}}Cl_{10^{23}-1}$. When one is dealing with condensed phases with infinite lattices, the removal or addition of single atoms produce such minute changes in the formula that one can have essentially minute and gradual changes in the properties of the phase as one changes its composition. Thus it is not surprising that condensed phases have ranges of composition and are not restricted to definite compositions or simple combining proportions. In fact, one can prove thermodynamically that no condensed phase at equilibrium can be restricted to a unique composition except at singular points such as a congruent melting or boiling point, peritectic and similar points, or at temperatures approaching absolute zero. Except for these singular points, condensed phases under all other equilibrium conditions must have appreciable ranges of composition or appreciable homogeneous ranges.

Consider the sodium chloride phase again. The phase which is in equilibrium with chlorine at 1 atm and therefore has a fugacity of chlorine of 1 atm cannot be the same sodium chloride phase which is in equilibrium with sodium metal and therefore has an activity of sodium metal close to unity. For we know that it is impossible to have sodium metal at unit activity and chlorine gas at 1 atm in equilibrium with one another. For the sodium chloride phase in equilibrium with sodium, one can from available thermodynamic data calculate the fugacity of chlorine corresponding to that phase. As one varies the composition from the sodium chloride composition in equilibrium with sodium to the sodium chloride composition in equilibrium with chlorine by adding chlorine atoms, the chlorine fugacity increases from a minute value to a value of 1 atm.

The changes in properties of sodium chloride across its homogeneous range are quite substantial. For example, its color changes from deep blue at the sodium-rich end to a white color at the chlorine-rich end (*1*). Likewise, its electrical and many other properties change quite markedly as one moves across the homogeneous range. For sodium chloride the range in composition is rather small, although it can be detected analytically without much difficulty. One

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finds that the homogeneous ranges will vary greatly from one phase to another. In some instances, a system of two components may correspond to a phase diagram with a complete homogeneous range from one component to the other. In other instances, there may be a number of intermediate phases, some of which have moderately wide homogeneous ranges and others which are fairly sharp homogeneous ranges.

It is important to recognize that the compositions at the limits of these phases would normally have no particular significance of their own. These limiting compositions will be different at each temperature and are not determined by the properties of the phase in question alone but are equally well determined by the properties of the phases that exist in equilibrium with the phase in question. Thus, under some circumstances, when it is possible by proper seeding to have a choice of phases which might be saturating a given phase, then the limit of homogeneous range will be different under the two conditions. The reader may consult the literature (*2*) to become familiar with the plots of free energy versus moles of one component per fixed amount of the other component for a series of phases which illustrate that the limits of the homogeneous ranges occur when one has a common tangent to two free energy curves corresponding to equal partial molal free energies for the two phases. Such plots clearly show the influence of the saturating phases upon the homogeneous range of a given phase.

Nomenclature Problems

It is quite customary to designate a phase by a formula which corresponds to some simple combining ratio. For example, the sodium chloride homogeneous range is referred to as the NaCl phase region; the iron (II) oxide homogeneous range, or the wüstite phase, is referred to as the FeO phase region. The use of these names does not imply any commitment about the range of compositions nor that the simple composition chosen to name the phase has any special significance within this phase region. Thus in the instance of the iron (II) oxide phase region, the composition FeO does not exist in an equilibrium system (*3*). Nevertheless it is useful to refer to this as the FeO phase region because of the fact that the X-ray pattern of compositions in this phase region corresponds to an ideal lattice of the sodium chloride type, which would have the composition FeO if it were an ideal lattice with no vacant lattice sites. Except at the absolute zero, it is not possible to have an ideal crystal for any substance at equilibrium and there will always be vacancies in the lattice. The vacancies in the cation sites need not be equal to those in the anion sites. In the instance of the wüstite phase, any attempt to equalize the vacancies in anion and cation sites would result in a phase with an iron activity greater than unity or a phase unstable with respect to disproportionation to metallic iron and an oxide phase richer in oxygen. Thus, even though we refer to phases such as the FeO phase, the TiO phase, the NaCl phase, etc., we do not imply by such a designation that the phase has a fixed composition corresponding to the ideal crystal structure. We make no other commitment than the designation of the crystallographic arrangement. For those who are accustomed to dealing with intermetallic compounds, this is quite obvious. However, many people who have dealt with nonmetallic compounds under rather restricted conditions often have not been aware of the importance of recognizing the existence of homogeneous ranges for all compounds.

There are many important respects in which not recognizing this can greatly handicap the understanding of chemical processes. Let us consider a few examples. Of course the very drastic change in electrical conductivity as a function of composition has been strongly emphasized in recent solid state work. However many of the other properties vary quite considerably.

As an example I might cite the cerium sulfide phase on which we worked some years ago (*4*). We had prepared a series of samples which according to the X-ray reports were identical. Within experimental error no difference could be detected in lattice contents. They all had the same structure, but their properties differed considerably. The color varied from black to red. The electrical conductivity varied from that of almost a metallic conductor to that of an insulator. The thermal conductivity likewise varied from high to very low. The thermal shock properties varied so markedly that the red sample behaved as a typical ceramic material that shattered very readily upon rapid heating or cooling whereas the black material could be heated 1000° per min. without shattering. The black material was an extremely good reducing agent whereas the red was an oxidizing agent under many conditions. The observation of a constant lattice parameter for the series of preparations is often taken as an indication of a very narrow homogeneous range, but in this instance the sulfur to cerium ratio varied from approximately 1.33 to 1.50. The ideal lattice corresponded to Ce_3S_4. Normally as one adds material to fill vacant sites one expects an expansion, so that as one adds cerium to the Ce_2S_3 one would expect an increase in the lattice parameter. However the cerium atoms with their extra electrons so greatly increase the binding of the lattice that the expansion that would normally be expected when holes are filled is almost exactly balanced by the increased strength of the bonds. Chemical analyses established that the $Ce_{3-x}S_4$ phase showed a range from $x = 0$ to $x = 0.3$ with only a slight change in lattice parameter.

Vaporization Processes

One of the topics which often causes a great deal of difficulty is the designation of a vapor pressure for a phase. When one recognizes the existence of a homogeneous range for a phase it is clear that there will be a range of vapor pressures as a function of composition. Unless one can characterize the phase in regard to its composition and can use a method of vapor pressure determination that does not appreciably alter the composition (so that one can associate the vapor pressure with a specific composition within a homogeneous range), the designation of a single vapor pressure for the phase region is normally meaningless. The phase will normally be saturated at either end of its range by some other phase. In a binary system at fixed temperature, the two condensed phases together with a gaseous phase constitute an invariant system with a unique characteristic vapor pressure. Thus if one does not measure

the vapor pressure as a function of composition in a homogeneous range, one must normally insure that a saturating condensed phase is present to characterize the system.

In most systems this means that one does not wish to use a "pure" sample which would correspond to some composition in the single phase region, as the system would not be uniquely characterized and the vapor pressure measurement would have no significance. One of the two phases which could saturate the phase in question must be added in order to produce a completely characterized system before measuring the vapor pressure. In some instances a constant boiling composition exists within the homogeneous range and if one uses a method of vapor pressure determination which allows the system to approach this constant boiling composition, there will be a third composition within the homogeneous range for which the vapor pressure measurement can be made without characterizing the composition of the phase. This generally does not occur. One must verify that such a condition is possible and insure that the method of determination allows one to reach this condition before attempting to make a measurement on a single phase system without carefully specifying the composition. In some systems the pressure may vary a millionfold across a homogeneous range, and it is quite meaningless to publish vapor pressures for a phase as is sometimes done unless one has made the system invariant by adding a saturating phase or unless one has insured constant boiling conditions. As the constant boiling composition will in general vary with temperature, one must be somewhat cautious in using the temperature coefficient of the vapor pressure to obtain heats of sublimation or vaporization as the heat may not correspond to any standard heat unless one has corrected for the changing composition of the constant boiling composition with temperature.

Thermodynamic Calculations

The prediction of chemical behavior by means of thermodynamic data can be seriously in error particularly if one does not take into account the homogeneous range of each phase. The thermodynamic data in the literature are normally given for one standard composition. The standard states may be hypothetical and need not correspond to any actual composition. In carrying out the calculations to predict actual behavior one must be prepared to calculate free energy changes for changes in composition from the standard state composition to the composition actually present at equilibrium. It is in this area that there is a most serious limitation to the use of thermodynamic data for prediction of chemical behavior, particularly in high temperature systems. This calculation of the free energy change in going from the standard composition to the actual equilibrium composition corresponds to knowing the activity coefficient as a function of composition within the homogeneous range. We have quite good data (*3*, *5*) for activity coefficients in the $Fe_{1-x}O$ homogeneous range and we are slowly accumulating data for the $TiO_{1\pm x}$ and UO_{2+x} homogeneous ranges (*6*), but among oxide systems the number of systems where even fragmentary activity coefficient data exist are extremely small. We know from the few examples that have been studied that the activity coefficients may vary in a complex manner in such homogeneous ranges. We will need much more data before we will be able to confidently predict activity coefficients in these homogeneous ranges and therefore reliably predict chemical equilibrium behavior.

Consider the example of the equilibrium between alumina solid and hydrogen gas. If the system were made invariant by addition of aluminum metal to convert the alumina to the composition which is saturated by aluminum metal, we could easily calculate the ratio of water to hydrogen partial pressures in equilibrium with the aluminum-rich alumina phase. However if the system had not been made invariant by addition of aluminum metal, the hydrogen-water ratio would be undefined unless we were to specify the composition of the alumina phase. Across the homogeneous range of the alumina phase, the water to hydrogen ratio will vary from an extremely high value at the oxygen-rich end to an extremely low value at the metal-rich end. Having specified the initial alumina composition and the relative amounts of hydrogen and alumina, as well as the hydrogen pressure, can we predict the water partial pressure that will result when equilibrium prevails? There are, of course, a number of choices of components that one can use to characterize the composition of the alumina phase, but let us take the simplest one, namely, the elements aluminum and oxygen as the components. To solve the problem we must know the variation of the activity coefficient of oxygen as a function of concentration in the alumina phase. Such data are not known at present to my knowledge, and there appear to be no adequate theories for confidently predicting such activity coefficients. Thus the particular problem of the equilibrium between hydrogen and alumina cannot be solved with the thermodynamic data now available. All that we can say is that, depending upon the relative initial amounts of hydrogen and alumina, the final equilibrium ratio (*7*) of water to hydrogen partial pressures will lie somewhere between 10^{10} and 10^{-14} at 1000°K. For each specific composition in the homogeneous range of the alumina phase there will be a specific value of the equilibrium water-hydrogen ratio somewhere between the limits indicated. Thus within the alumina homogeneous range we have a variation from an extremely powerful reducing agent to an extremely powerful oxidizing agent. It is not surprising that other properties likewise show considerable variation as a function of composition.

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(5) Darken, L. S. and Gurry, R. W., *J. Am. Chem. Soc.*, **68**, 798–816 (1946).
(6) Mah, A. D., et al., Bureau of Mines Report of Investigations, 5316 (1957).
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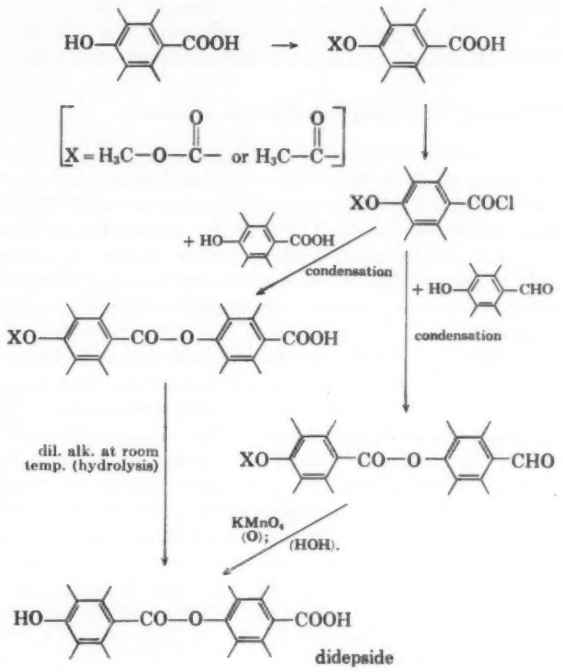
Depsides: a Synthesis by Emil Fischer

The annual meeting of the German Natural Scientists and Physicians was held in 1913 at Vienna. The highlight of the meeting was Emil Fischer's lecture on his investigations[1] on the "Synthesis of depsides, lichen substances and tannins." During the introductory remarks Fischer observed that phenol carboxylic acids possess "the ability to form ester-like anhydrides with one another in such a manner that the carboxyl of the first molecule unites with the phenol group of the second" and continues "such ester-like anhydrides, Freudenberg[2] and I have named 'depsides.'" This was accomplished exactly fifty years ago, in 1910.[3] Fischer introduced this term "depside" from the Greek word δέψειν (to tan), since a typical depside digallic acid occurs as a part of the molecule of tannin from gallnuts. Also the name depside emphasizes the analogy with polypeptides derived from simple amino acids and polysaccharides from simpler sugars—the two fields that were enriched by Fischer's fundamental contributions. Based on the number of phenol carboxylic acid units present, a given depside is named di-, tri- or tetra-depside.



Synthesis of Depsides

In a phenol carboxylic acid molecule, the hydroxyl groups are first protected by carbomethoxylation, and the carboxyl group is then activated by conversion to the acid chloride. The latter is allowed to react with a second molecule of free hydroxy acid when the hydroxyl para to the carboxyl group preferentially reacts forming the ester linkage between the two molecules. Mild hydrolysis of the product selectively eliminates the carbomethoxy groups yielding the free didepside. Fischer later found that acetylation was another convenient method of protecting the hydroxyl groups.

An alternate method of synthesis developed by Fischer in collaboration with his son, H. O. L. Fischer,[4] involves the condensation of the carbomethoxy acid chloride with a hydroxy aldehyde and then oxidation of the aldehyde to the carboxyl group. By suitable modifications in his methods, Fischer also achieved the synthesis of ortho didepsides.

Actually it was his researches in the field of polypeptides that induced Fischer to take up the synthesis of depsides. A necessary intermediate in the synthesis of glycyltyrosylglycine is the acid chloride of chloracetyltyrosine. During the preparation of the latter, since a free phenolic hydroxyl interferes with the action of phosphorous pentachloride, Fischer "thought of the scheme of protecting the latter (phenolic hydroxyl group) by introducing a group which subsequently could be easily removed, and chose for this purpose the carbomethoxy group." "The application of this process to the common phenol carbonic acids is the starting point for" the synthesis of various depsides.

Fischer investigated the versatility of the carbomethoxylation technique. He made a detailed study of the action of methyl chloroformate on various phenol carboxylic acids wherein the carboxyl is attached either to the benzene ring or to a side chain. Fischer also developed the method for the easy elimination, when required, of the carbomethoxy group. Likewise, he made a systematic study of the conversion of carbomethoxy acids into their acid chlorides using phosphorous pentachloride and explored the potentialities of these acid chlorides in organic syntheses; one of such uses proved to be in the synthesis of depsides.

Diprotocatechuic acid, *o*-diorsellinic acid, di-*o*-coumaric acid, di-β-hydroxynaphthoic acid, α-hydroxynaphthoyl-*p*-hydroxybenzoic acid, and vanilloyl vanillin are some of the numerous didepsides synthesized by Fischer and his collaborators.

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[2] For biography see Oesper, R. E., J. Chem. Educ., **28**, 426–27 (1951); Helferich, B., *Angew. Chem.*, **68**, 81–84 (1956); Anon., *Proc. Chem. Soc.*, 97–98 (1959).

[3] Fischer, E., and Freudenberg, K., *Ann.*, **372**, 35 (1910).

[4] For biography see Anon, *Chem. Eng. News*, **27**, 1437, 1495 (1949); Anon, *Chem. Eng. News*, **27**, 2070 (1949).

Properties of depsides. By the action of diazomethane both the carboxyl and hydroxyl groups of a depside get methylated resulting in fully methylated esters, which are beautifully crystalline and melt without decomposition, unlike the free depsides. Consequently, these derivatives are very useful in characterizing the depsides. Some of the depsides are precipitated by glue solutions and form precipitates with quinine acetate, thus showing their similarity to tannins.

Tri and tetra depsides. Fischer extended his synthetic routes to tri- and tetra-depsides also. Vanilloyl-*p*-hydroxybenzoyl-*p*-hydroxybenzoic acid and tri-*p*-hydroxybenzoyl-*p*-hydroxybenzoic acid are examples.

Naturally-Occurring Depsides and Tannins

In nature, depsides occur in lichens, and the most common of these depsides is *p*-diorsellinic acid or lecanoric acid. Fischer made an exhaustive study of this compound—put forth evidence for its structure, synthesized it and proved the identity of the synthetic and natural samples. He also carried out a detailed examination of evernic acid, another naturally occurring depside.

Fischer's researches on depsides led to his investigations on the more complex products presented by nature, the tannins—high molecular-weight substances built up from depsides and sugars. He made a thorough study of tannin—the tanning substance of gallnuts. These investigations have culminated in his synthesis of penta (*m*-digalloyl)-β-glucose in collaboration with Bergmann[5] in 1918. It was found that this artificial tannin had a very close resemblance with the principal constituent of Chinese Tannin (from gallnuts) in most of its properties. The resemblance was so much that Forster[6] states that "it is therefore justifiable to claim that gallotannic acid or gallnut tannin has been synthesized."

Conclusion

Throughout his researches, Fischer gave due credit to his associates. This is best expressed in his own words:

> I have, in this investigation, enjoyed the valuable assistance of different young professional colleagues, whose share is evident from the titles of the single publications, and of these I will mention particularly Mr. (now Prof.) Karl Freudenberg.

No amount of appreciation can parallel the monumental achievements of Emil Fischer in synthetic organic chemistry, in particular in the fields of carbohydrates, proteins and purines—fields of immense biological significance. From the vast amount of accumulated literature on Fischer, it is rather difficult to choose which words of admiration are the best. The author feels that the pen of Arthur B. Lamb[7] is most striking in this regard. Reviewing the collected works of Emil Fischer, he says:

> They make us realize not only the genius but also the virility and fecundity of Fischer's mind. Emil Fischer surely approximated very nearly the ideal investigator. His discoveries were not accidental. He selected broad problems of capital importance, and was not dismayed by their difficulty. He attacked them logically and aggressively. His success was indeed marvelous and mankind owes him an incalculable debt of gratitude.

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[5] For biography see Anon, *Chem. Eng. News*, **22**, 2059 (1944); NEUBERGER, A., *Nature*, **155**, 419–20 (1945); HARINGTON, C. R., *J. Chem. Soc.*, 716–18 (1945); CLARKE, H. T., *Science*, **102**, 168–70 (1945); MCLAUGHLIN, G. D., *J. Am. Leather Chem. Assoc.*, **40**, 2–3 (1945).

[6] FORSTER, M. O., "Emil Fischer Memorial Lecture," *J. Chem. Soc.*, 1178–79 (1920); For Forster's biography see Anon, *Curr. Sci.*, **1**, 302 (1933); Anon, *Curr. Sci.*, **14**, 141–42 (1945); SIMONSEN, J. L., *Nature*, **156**, 13–14 (1945); ARMSTRONG, E. F., AND SIMONSEN, J. L., *J. Chem. Soc.*, 550–57 (1946).

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Science News for Students

Wesleyan University (Middletown, Connecticut) announces *Science and Math Weekly*, a new weekly science newspaper for senior high school classes in biology, chemistry, physics, and mathematics. *Science and Math Weekly* (for senior high schools) is a companion publication to *Current Science* (for junior high grades), in the American Education Publications' weekly series.

The new weekly answers the demand of many teachers for bridging the gap between the textbooks and recent developments in science and science instruction. (EDITOR'S NOTE: We congratulate the editors and recommend that readers of THIS JOURNAL investigate their students' reactions to this publication.) The editorial staff at Wesleyan combines the efforts and experience of college and university teachers with high school teachers.

Science and Math Weekly is edited for high school classes and is also valuable for use in junior college science courses. Scheduled for publication are 32 issues during the school year. First issue appeared December 7, 1960. The cost per student is 50 cents per semester, $1.00 per school year, in class orders of 10 or more copies. The teacher receives a free desk copy and the Teacher's Editions with class orders. Subscriptions and inquiries may be sent to *Science and Math Weekly*, Education Center, Columbus 16, Ohio.

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Using Filmed Demonstrations in General Chemistry

Can special films be used in place of actual demonstrations during lectures? If this procedure produces results with students, it may suggest considerable savings in time and money. The experiment here outlined sought to answer the question.[1]

The experiment was conducted in a first semester general chemistry course at the Ogontz Campus of the Pennsylvania State University. The 47 students enrolled in the course were assigned by randomization to an experimental group of 23 and a control group of 24 subjects. The first comprised 14 first semester freshmen and 9 second semester freshmen; the second, 16 first semester freshman and 8 second semester students. The students represented various curricula, predominately engineering, agriculture, and chemistry.

Eighteen 30-min kinescopic recordings (16 mm films made from television presentations) which had been prepared by the Pennsylvania State University[2] for the Department of Defense was used in about half of the lectures that were given to the experimental group. These films heavily emphasized demonstrations. The control group received the usual classroom lecture demonstrations throughout the semester. The course in which the experiment was performed is a five credit course comprising two 50-min lectures, two 50-min recitations, and a 3-hr and 50-min laboratory each week. Both groups were taught by the author and received the same recitation and laboratory periods.

The subject matter covered by the series of 30-min films is summarized as follows:

(1) Kinds of substances; demonstrations of solidification and distillation.
(2) Fundamental particles; showed Crookes tubes, Wilson cloud chamber, and the Geiger counter.
(3) Elements and atoms; laws of chemical combination.
(4) Compounds and formulas; empirical, molecular, and structural formulas, and energy changes accompanying chemical reactions.
(5) Composition of the atmosphere; liquid nitrogen.
(6) Structures and properties of carbon and its compounds; models were shown.
(7) Behavior of gases.
(8) Kinetic theory of gases; diffusion, velocity, and condensation.
(9) Liquids and solids; vapor pressure of various liquids.
(10) Periodic classification of the elements; relationship of properties of the elements to their positions.
(11) Nuclear structure of atoms; Rutherford's experiment showing atoms contain very small, heavy nuclei.
(12) Electronic structures; energy levels and ionization potentials; models.
(13) Electronic structure and chemical bonding; behavior of covalent and ionic solids.
(14) Chemical bonds; formation of ionic and covalent substances.
(15) Chemical reactions; electrolysis, oxidation-reduction half reactions.
(16) Solutions; concentrations.
(17) Chemical equilibrium.
(18) Ionic equilibria; the common ion effect, complex ions, and dissolution of slightly soluble substances.

Three 50-min tests and a final examination were used to evaluate the experiment. These tests were of the multiple choice type composed of a word or sentence choice, sentence completion, equations, and problems consisting of empirical and true molecular formulas, weight-weight and weight-volume, freezing point depression and boiling point elevation, Faraday, equilibrium constant, and specific heat.

The final results are summarized in the table below.

Results of Chemistry Film Experiment

Exams	Means: film-lecture group	Means: control group	F	Significance
I	68.40[a]	63.66[a]	1.05	0.05
II	63.38	66.79	0.02	0.05
III	60.50[a]	58.43[a]	3.23	0.05
Final	49.29	48.95	0.10	0.05

[a] Mean adjusted by covariance analysis.

The data in the table indicate that students who saw the filmed lecture-demonstrations performed as well on tests as those who experienced live lecture-demonstrations. A summary evaluation of student opinions was generally favorable. They appreciated the advantage of films for carefully planned coverage of more material. The greatest objection was that the presentation was too rapid. Repetition, interruption for questions, or better introduction are techniques to be considered for overcoming this objection in the future.

General Conclusions

Filmed lecture-demonstrations in college chemistry can be used at least as a partial substitute for live lecture-demonstrations.

This finding has implications for courses in industry, in the services, or in some colleges where a sufficient number of experienced chemistry lecturers may not be available. Regular use of such films demands excellent projection facilities and careful planning to fit them into the whole classroom program.

Short films of chemistry demonstrations would be a valuable resource to use in live lectures. Such films could cover demonstrations which take a long time to prepare or those that are difficult to perform reliably in the lecture room due to lack of laboratory controls.

[1] The experiment was conducted in conjunction with the Division of Academic Research and Services of the Pennsylvania State University.

[2] The presentations were made by Dr. C. G. Haas, Jr., Pennsylvania State University, University Park, Pennsylvania.

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A Motor-Driven Variable Transformer

There are a number of applications in the laboratory where a device providing a uniform stepless heating rate would be desirable. This is particularly true in studies using the thermal techniques of differential thermal analysis (DTA) (*1*) and thermogravimetric analysis (TGA) (*2*). The interpretation of data from both DTA and TGA is generally rendered less ambiguous when a controlled linear temperature rise prevails during an experimental determination. Furthermore, the results of different workers are best compared under the same heating program. Manual adjustment of the temperature rise of a furnace has frequently been employed through use of an autotransformer to regulate the power input to the heating elements (*3*). Thus, it is the intent of this note to present the details of construction of a motor driven variable transformer which is of simple design, incorporates readily available parts, and allows for a wide choice of essentially linear heating rates.

A schematic drawing of the assembled apparatus is given in Figure 1. The bottom end of the center shaft of a variable transformer[1] was coupled directly to the drive-shaft of a synchronous motor of desired rotation rate.[2] Torque was applied to the transformer shaft by the motor as the motor frame lodged against one of the three wooden support rods.

Figure 2 shows a representative series of heating rate curves as functions of the rotation rate of the motor shaft. The measurements were made in a small muffle furnace (cavity size: 4 in. × 4 in. × 5 in.) using nichrome wire heating elements drawing about 8 amp at 110 v. The temperature signal from a chromel-alumel thermocouple was continuously plotted as a function of time on a potentiometric recorder. It is usually necessary to start a program at some transformer setting above 0 v in order to achieve a linear heating rate within a short time interval. This initial setting and the actual heating rate obtained at any given drive-shaft speed will be dependent on the configuration of the furnace assembly used.

[1] Powerstat, Type 3PN-116, 0–140 v, 7½ amp, the Superior Electric Co., Bristol, Conn.

[2] Synchron Timing Motors, obtained from Herbach and Rademan, Inc., 1204 Arch St., Phila. 7, Pa.; catalogue no. HI-34 (½ RPH), HI-35 (⅓ RPH), HI-37 (⅙ RPH), HI-39 (1/12 RPH), and HI-40 (1/24 RPH).

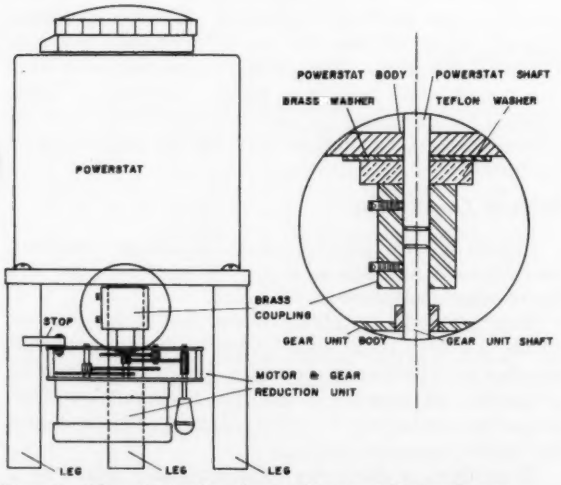

Figure 1. Schematic drawing of motor-driven variable transformer.

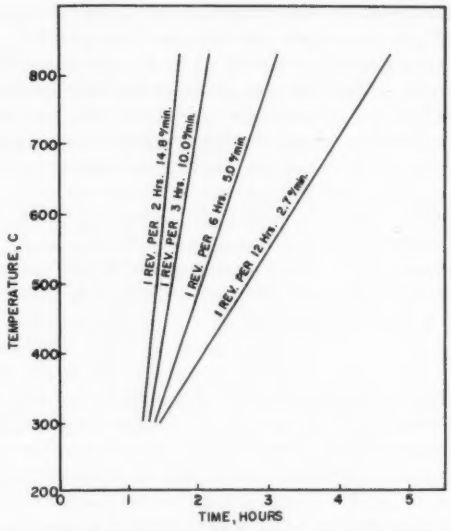

Figure 2. Representative heating rate curves as functions of motor drive-shaft rotation rate.

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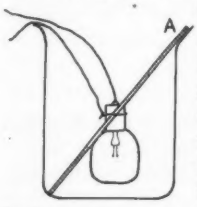
A Flashbulb Bomb Calorimeter

This report describes a do-it-yourself experiment in thermochemistry using Sylvania Blue Dot M-2 flashbulbs as calorimeter "bombs." As compensation for the lack of accuracy and precision, the student gains an intimate acquaintance with the working elements of a calorimeter; a demonstration experiment with standard equipment may be less satisfying in this respect. In addition, certain chemical complications involved in the determination of a heat of formation are emphasized.

The flashbulbs are filled with almost pure aluminum (99.00% minimum) in an atmosphere of pure oxygen. The flash begins when a fine tungsten filament, heated to incandescence, ignites primer beads of zirconium, potassium perchlorate, and a binder at the ends of the lead wires. The primer beads then generate a heat stream sufficient to ignite the shredded aluminum. The combustion lasts about $^1/_{30}$ sec and peak brilliance is reached at 13 millisec. Although a minimum of 3 v is specified for firing, a large ignition dry cell with 1.5 v was used and found satisfactory.

The calorimeter is made by bending a layer of pasteboard around a 150 ml beaker just below the lip and securing it with tape. A second layer of pasteboard around the beaker extends to the top of the lip. Then two pasteboard circles are cut out. One is reserved for the top of the calorimeter and the other is taped to the bottom; masking tape is preferred.



Bomb assembly.

A length of copper wire (20 gauge) is soldered to the soft metal tip of the flashbulb base. A second piece of wire is tightly twisted around both the bulb base and a wooden splint as shown in the figure. The second wire piece thus serves not only as a second electrical lead but also to secure the "bomb" within the calorimeter. The bulb base is apparently an aluminum alloy and ordinary solder cannot be used to fasten the second lead. To insure good electrical contact, the metal surfaces should be clean, and it may be necessary to remove the thin plastic film partially covering the bulb base. A piece of Scotch tape applied to the splint and beaker lip at *A* is sufficient to hold the bomb assembly steadily in place.

The calorimeter is completed by pipetting 100 ml of distilled water into the beaker and fitting a stirrer and thermometer (0.1 or 0.2 degree divisions) through the pasteboard top which is subsequently taped down; cork borers are used to cut holes. The stirrer, made from 4 or 5 mm solid glass rod, has a single right-angle bend at the bottom with the tip flattened into a paddle shape.

After firing, the maximum temperature rise is noted; the usual extrapolation to zero time is not employed. The student must then dissect the calorimeter, determine a value for the heat capacity, and calculate the energy evolved in combustion.

The heat capacity of each item in the calorimeter assembly is determined by multiplying its weight by its specific heat. The M-2 bulb was broken into its metal and glass components which were weighed, giving an estimated heat capacity of 0.8 cal/deg. A value of 1.0 cal/deg was assumed for the thermometer. As shown in the table the student calorimeter has a total heat capacity of about 113 cal/deg. The heat produced by the flash reaction can then be determined from the temperature increment which should be about 0.80 degree centigrade.

More accurate experiments using a Beckmann thermometer in a Dewar flask of known heat capacity showed that the M-2 flashbulb developed 88.1 cal; an average deviation of 1.8 cal. was observed. Results obtained with the student calorimeter were in good agreement with this value.

The enthalpy change (−200 kcal) for the reaction

$$Al_{(s)} + {}^3/_4\, O_{2(g)} \rightarrow {}^1/_2\, Al_2O_{3(s)}$$

is related to the energy change in the bomb (per gram-atom of Al) by the equation

$$\Delta H = \Delta E + \Delta nRT$$

or

$$\Delta E = \Delta H + {}^3/_4\, RT$$

At 25° the last term amounts to only 0.45 kcal. A consideration of the precision of the experiment shows that

Calculated Heat Capacity of Calorimeter at 25°

Item	Weight (g)	C_p (cal/g-deg)	Heat Capacity (cal/deg)
Water	99.7	0.998	99.5
Beaker	56.0	0.20[a]	11.2
Stirrer	3.0	0.20[a]	0.6
Copper leads	1.3	0.093	0.1
Thermometer	...	...	1.0
M-2 Flashbulb	0.33[b]	0.22	0.1
	3.50[c]	0.20	0.7
		Total	113.2 cal/deg

[a] Reported for Pyrex brand glass.
[b] Aluminum alloy.
[c] Glass and plastic.

The author wishes to thank Mr. D. E. Armstrong of Sylvania Electric Products Inc. for details of flashbulb construction.

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the difference between ΔH and ΔE is not significant so that ΔE is taken to be -200 kcal/mole. A comparison of the heat of the flashbulb reaction with ΔE shows that 11.9 mg of aluminum is consumed. Given this value, the student can measure the heat produced in his calorimeter and perform the above calculations in reverse, obtaining the heat of combustion of aluminum at constant volume and constant pressure. Finally, the latter value can be doubled giving a value for $\Delta H°_f$ of $Al_2O_{3(s)}$.

Aluminum oxide, corundum, or alumina as it is variously called, exists in several crystalline modifications. There is some ambiguity in a thermodynamic statement which gives no specification. It is found by x-ray diffraction that when aluminum is burned α-corundum is formed along with a type designated x-Al_2O_3. The enthalpy of transition for the conversion of x-Al_2O_3 to α-Al_2O_3 is about -10 kcal/mole, therefore doubling the heat of combustion of Al does not give the heat of formation of α-corundum; the necessary corrections give a final value[2] of about -402 kcal/mole. Another low temperature modification, γ-Al_2O_3, has a less negative heat of formation than α-Al_2O_3 and is easily converted to the latter by application of heat. For this reason (presumably) it is not formed in combustion.

Since the M-2 flashlamp contains about 16 mg of Al foil, combustion is only about 75% complete owing to a lack of oxygen. Indeed, an excess of oxygen would lead to prohibitively high pressures at the high temperature attained during burning. The excess aluminum melts during the flash and is splattered against the interior walls of the bulb. The P-25 (press) bulb, which contains about 33 mg of Al, can also be used as a bomb. Its larger volume makes it somewhat less convenient to handle; a 200 ml beaker is recommended. Its heat capacity is about 1.5 cal/deg. and it liberates about 190 cal of heat showing that 26 mg of aluminum is consumed.

Recently (1958) zirconium foil has been introduced as a flashbulb filler. Smaller than a peanut, the Zr-filled Sylvania AG-1 supplies about the same amount of light as the M-2 which is four times its size. In general, zirconium burns with a light output 100% greater than aluminum with less pressure at the flash peak. The enthalpy of formation[3] of $ZrO_{2(s)}$ is -258 kcal/mole whereas the value for $^1/_2$ mole of $Al_2O_{3(s)}$ is -200 kcal. Hence, on an equal weight basis zirconium liberates much less heat than aluminum does.

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[3] Thermodynamic data are taken from LATIMER, WENDELL M., "The Oxidation States of the Elements and Their Potentials in Aqueous Solutions," 2nd ed., Prentice-Hall, Inc., New York, **1952**, pp. 270 and 282.

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Faraday's Laws in One Equation

Examination of 62 general chemistry texts (high school and college) and 11 physical chemistry texts at the book exhibit of the September 1959 ACS meeting showed that many are still presenting Faraday's laws of electrolysis virtually in the original form.[2] Some are replacing them with the statement that 96,500 coulombs (most recent value 96,489.9 ± 2.0) passed through an electrolytic cell, will cause one gram equivalent weight of material to react or form at an electrode. The advantages of this presentation are recognized by many teachers. The following simple derivations of the single-equation statement, along the lines suggested by Noyes and Sherrill,[3] may have interest.

Analysis of the two laws reveals that they concern three variables: quantity of electricity (charge, q), amount of material (mass, m), and gram equivalent weight (m_e). In each law, one variable is held constant. The first law states that q is proportional to m when m_e is constant, i.e., a particular substance in a particular cell is being considered. The second law states that if q is constant, m is proportional to m_e.

A single proportionality which includes both statements is:

$$q \propto \frac{m}{m_e} \tag{1}$$

since, if m_e is constant, $q \propto m$, and, if q is constant, m/m_e = constant or $m \propto m_e$. Rewriting (1) as an equation:

$$q = \frac{m}{m_e} \times (\text{constant}) \tag{2}$$

Calling this constant F (for Faraday) and m/m_e the number of equivalents, n:

$$q = nF \tag{3}$$

is a mathematical statement of Faraday's law.

This form of Faraday's law, with the definitions

$$n = \frac{m}{m_e} \quad \text{and} \quad q = \int_{t_1}^{t_2} I\,dt$$

can be used to solve readily all the general chemistry problems commonly given on Faraday's laws.

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Proceedings of the
California Association of Chemistry Teachers

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Antecedents to Modern Concepts of Configurational Symmetry in Chemistry

Many students of chemistry are apt to regard the assignment of such geometrical forms as the tetrahedron and octahedron to molecules and ions as a triumphant union of modern mathematics and twentieth century chemistry. Unfortunately, such a conclusion ignores the fact that the application of geometric shapes to various scientific concepts is one of the oldest practices in the intellectual heritage of man. In order to give students the opportunity to acknowledge and appreciate the indebtedness which we all owe to the ancient natural philosophers, this paper seeks to review the philosophical background which is the basis for modern spatial chemistry.

One of the perennial and unifying abstractions in the entire history of ideas is the notion of symmetry. Symmetry implies an equal correspondence in size, shape, and relative position of parts of a whole as distributed about a median point, line, or plane. The presence of symmetry is associated with balance, proportion, harmony, and beauty. The importance of this subject to many fields of intellectual endeavor has been emphasized by various writers. The eminent historian of science, George Sarton (*1*), has said of the role played in science by symmetry: "[it] has always and will always remain one of the safest guides . . . of the scientist." Among the classics in the field we may mention briefly the following: Pierre Curie wrote a number of memoirs on the extension of the concept of symmetry from the field of crystallography to that of physics (*2*). He arrived at generalities which were summarized very succinctly by Marie Curie (*3*). A treatment of symmetry with emphasis on chemistry has been given by Jaeger (*4*). The important role of symmetry in deducing the mathematical relationships in organic nature has been demonstrated by Thompson (*5*). Herman Weyl, who was one of this century's leading mathematical physicists and philosophical mathematicians, has presented a superb integration of art and science through the principles of symmetry (*6*).

It might seem that as science becomes more mathematical and less dependent on mechanical models with various forms, that symmetry would diminish in importance. But this is not so. Regarding one of the most fundamental and abstract branches of modern science, theoretical physics, we find a recent writer (*7*) declaring that symmetry is one of the elements that pervades the logical foundations, the physical content, and the mathematics of almost the whole field. The recent jarring discovery of the nonconservation of parity has once again projected symmetry to the forefront in the philosophy of physics (*8*).

The relations between number and symmetry became known in a primitive way by early man when, for instance, he found that three similar objects taken as the spoils of war could not be divided equally among two warriors. Similar relations were known also as a matter of every day practice by the tradesmen and others of early civilization. However, among the first to formalize the principles of symmetry and numbers was Pythagoras (*ca.* 582–500 B.C.). He pointed out the important distinctions between odd and even numbers and classified numbers as triangular and square. The connection between geometric form and number became evident because the Greeks at that time had no numerals, so they represented the number of objects by an equal number of pebbles placed on the ground. Some numbers, made from pebbles equally spaced from their nearest neighbors, were of triangular shape (3, 6, 10). Other similarly spaced groups of pebbles (4, 9, 16) assumed the form of squares.

The ancient natural philosophers did not limit their study of symmetry to two dimensions. The sphere, because of its perfection of rotational symmetry, was admired for itself and was utilized to satisfy the desire of paying homage to the grandeur of the universe. The Pythagoreans regarded the sphere as the most excellent three-dimensional figure. Aristotle assigned sphericity to the heavenly bodies as a tribute to the perfection of their sublime nature. Although the sphere occupied the position of greatest esteem from the standpoint of symmetry and was perhaps the easiest solid figure to comprehend, other geometric forms also became the objects of study, particularly the five regular polyhedra. These are the cube, tetrahedron, octahedron, icosahedron, and dodecahedron, bounded respectively by the following planes: six squares, four equilateral triangles, 20 equilateral triangles, and 12 regular pentagons. (The areas of the planes bounding a given figure must be equal.) The tradition of the discovery of these figures is rather confused (*9*). Of

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course, it is fruitless to try to designate the first recognition or construction of the cube or tetrahedron. As for the origins of the other three, there is some difference of opinion. Heath (*10*) and Sarton (*11*) believe that the earliest Pythagoreans were acquainted with all five of these polyhedra. Burnet, on the basis of the scholia to Euclid as given by Heiberg, claims there is reason to believe that they were familiar only with the cube, tetrahedron, and dodecahedron (*12*).

An interesting aspect of the history of the five regular polyhedra is the manner of construction, i.e., whether the figures were put together manually or whether they were deduced theoretically. The first Pythagoreans did not know enough geometry for the latter, but those figures with which they were familiar were made by mechanically joining appropriate numbers of plane figures. However, a later Pythagorean, Theaetetus (*ca.* 415–*ca.* 369) was the first to write a complete and systematic treatise on the five regular solids, and he is generally given credit not only for the theoretical constructions (*13*), but also for proving that there can be no more than five such solid figures (*14*). The crowning conclusion of the famous "Elements" of Euclid (*ca.* 300 B.C.) is the presentation of the rigorous theoretical construction of these solids and the determination of the relation of the edge of a solid to the radius of a circumscribing sphere (*15*). Euclid owes much of the substance of his proofs to Theaetetus.

Thus far we have been considering symmetry and the five regular solids from the standpoint of pure mathematics. Now we must examine the origins of the extension of these ideas to the properties of matter. After Empedocles (*ca.* 490–*ca.* 432 B.C.) put forth his theory of the four elements (air, earth, fire, and water), the Pythagoreans of the time adopted his theory by assigning one of their favorite figures to each element. Earth is composed of cubes, fire of tetrahedra, air of octahedra, and water of icosahedra. The dodecahedron was associated with the divine creation of the universe as a whole.

The Pythagorean theory was presented by Plato in the form in which it appears in his "Timaeus" (*16*). In this dialogue, Plato proposes that the four elements are present in the world as a whole in certain proportions, but they may be distributed in an infinite variety of ways to give the many observed substances. Also, the faces of the elemental shapes may be of different sizes, and these further allow for the great multiplicity of bodies. With regard to penetrability and mobility, the elements obviously rank in the order, fire, air, water, and earth. Hence fire must be composed of tetrahedra, with sharp points; air and water must be made of particles with points of lesser sharpness, namely, octahedra and icosahedra, respectively. These three elements are interconvertible. By the action of appropriate forces, figures of a given type can be dismantled, into their equilateral triangles (or proportional subdivisions thereof), and then reassembled into a different shape. Earth, in contrast, was stable and immobile, and hence must be made of cubes. Although cubes can be rendered asunder into their square planes, the latter can recombine only to give the original earth. Hence, the latter is incapable of transformation, and neither fire, air, nor water can be converted to earth, because their triangles cannot be formed into a cube. On the basis of this theory, Plato offered ingenious explanations for a number of transformations, such as the change of ordinary water into its other two states; fusion, solidification, and corrosion of metals; combustion, solubility, etc.

Although there are divergent opinions about the scientific attitude of Plato and the effect of his thought on later science (*17*), nobody denies that the "Timaeus" exerted an enormous influence for centuries on subsequent formulations of the philosophy of nature. Its Greek, Latin, and Arabic traditions are outlined by Sarton (*18*). In his introduction to this dialogue, Jowett (*19*) says:

> Of all the writings of Plato the "Timaeus" is the most obscure and repulsive to the modern reader, and has nevertheless had the greatest influence over the ancient and medieval world.

Stillman (*20*) says:

> The theories of Plato, as expressed principally in his "Timaeus," while contributing little of permanent value to science, exerted a great influence upon ancient and medieval notions of matter and its changes . . .

The tradition of Plato was carried on by philosophers and mathematicians, not only in the Academy (which lasted until 529 A.D.), but in the various intellectual centers of the world. The rise of alchemy provided another vehicle of transmission of Plato's ideas, for his speculations on the interconvertibility of the elements provided a theoretical basis for the transmutations of the alchemists. The writings of the early alchemists recognize Plato as their guide and doctrinal authority (*21*), and traces of his notions can be found in alchemical writings even as late as the 17th century (*22*).

In the first half of the Middle Ages, Plato was regarded as the supreme authority in philosophy. As expressed succinctly by Shorey (*23*):

> The shortest cut to the study of the philosophy of the early Middle Ages is to commit the "Timaeus" to memory. . . . The medieval writers paraphrased and glossed and quoted the "Timaeus" endlessly . . . The Platonic philosophy of the four elements was the basis of their physics and chemistry and they observed its resemblance to the atomic theories of Democritus . . .

Later in the Middle Ages there was a decline in the study of Plato, followed by a modest revival at the School of Chartres (*24*) with the "Timaeus" occupying a large share of attention, and finally almost a total eclipse by a resurgent Aristotelianism. However, with the Renaissance, and especially with the Platonic Academy of Florence, Plato's influence reached its zenith. By the middle of the 15th century, the symmetry of the five regular polyhedra was stimulating the imaginations not only of philosophers and mathematicians, but of artists as well (*25*).

The grip which Plato's mysticism could exert over the minds of men of the highest intellectual caliber is illustrated by Kepler (1571–1630), on the very eve of the scientific revolution. While still a young mathematics lecturer, his interest in astronomy challenged him to find the reasons behind the numbers and sizes of the then known planets. Failing to discover any single mathematical rule, he resorted to geometry, placing the five regular figures within the spaces of the six spherical planetary orbits. Thus the six spheres of diminishing sizes, Saturn, Jupiter, Mars, Earth, Venus,

and Mercury, are separated, respectively, by the cube, tetrahedron, dodecahedron, octahedron, and icosahedron. With this device Kepler found that the radii of these spheres harmonize approximately with the average distance between the earth and the five other planets. This scheme is represented by Kepler with a large plate in his earliest work on cosmology (*26*). This episode in the history of science, though a product of fancy, is important as a vehicle of transmission of the concept of Platonic symmetry. Kepler placed the five regular polyhedra within the framework of early modern science. His prestige as an astronomical genius kept these figures from possible rejection and disuse, a fate suffered by many concepts of Greek origin.

Just a few years after Kepler's death, Platonism manifested itself in the writings of William Davidson (*ca.* 1593–*ca.* 1669), a chemist who did much to assist the transformation of alchemy to chemistry (*27*). He gave public instruction in chemistry at Paris, and for his students he published in 1633–35, a practical and theoretical text, "Philosophia Pyrotechnia," in four parts. Davidson was steeped in the Platonic precepts of number, proportion, and geometrical symmetry, and these are very prominent in the theoretical sections of his book. For instance, in the French edition of 1675 (*28*) he devoted a whole chapter to the regular solids. They are portrayed most importantly, and are made the basis of numerous lengthy and fantastic elaborations linking their characteristics with chemical properties and principles.

Chemistry matured gradually in the 17th century by purging itself of mystical alchemy, but the concept of symmetry of the regular solids, freed from Platonic implications, persisted in the minds of later chemists. Thus Dalton, commenting on some work of Berzelius, suggests that oxygen atoms may be regular tetrahedra (*29*). It was Wollaston, almost simultaneously with the postulation of the Daltonian atom, who predicted stereochemistry. Suggesting that the characterization of a compound could not be accomplished merely by knowing the relative numbers of combined atoms, he said (*30*), "we shall be obliged to acquire a geometric conception of their relative arrangement in all three dimensions of solid extension." His forecast came to fruition in the work of Pasteur, Le Bel, van't Hoff, Wislicenus, and Werner. Today, even in elementary courses in chemistry, the cube, tetrahedron, and octahedron are presented as the symmetrical basis for a large portion of ionic and molecular architecture.

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NSF Summer Institutes in Chemistry: Addendum. . .

Adelphia College, Garden City, L. I., New York, as it has for the last three years, will once again have a Summer Institute for high school teachers of chemistry. Through some oversight, no mention is made of Adelphia College in the listing in this Journal, January 1961, p. 35. The subject matter of the institute deals mainly with bonding theory and modern instrumental methods of chemistry. Applications for attendance may be obtained from Dr. Howard A. Robinson at Adelphi College.

LETTERS

To the Editor:

In his letter published in THIS JOURNAL, **37,** 549 (1960), Professor Keighton of Swarthmore College criticized the omission of that institution from the list of liberal arts colleges whose faculty had published articles abstracted in Chemical Abstracts [See THIS JOURNAL, **37,** 316 (1960)]. I state again that my classification was based on the designations in the 9th edition of the "College Blue Book." The omission of an institution which includes a school of engineering seems consistent. The standing of any such professional school would be jeopardized by failure to publish in recognized scientific journals. Very seldom is the same true of liberal arts colleges. Not only is it often possible for professional schools to secure funds for research, but they often employ faculty members who give their full time to that activity. Again, such a condition is seldom encountered in liberal arts colleges.

I regret that the adoption of this criterion may have resulted in the omission of worthy institutions. Their omission did not imply the lack of productivity in research.

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To the Editor:

A recent article by Ricketts in THIS JOURNAL, **37,** 311 (1960), discussed an experimental approach to qualitative analysis as an exercise in deductive reasoning. We have used a similar approach as a way of doing qualitative analysis in our terminal chemistry course.

Students are assigned six cations and two sets of test reagents. The cations are chosen so that the separations involved will illustrate basic principles of chemical equilibria. The primary test reagents are used on separate 0.5 ml portions of each cation. Where precipitates form, secondary tests are carried out on separate portions of the precipitates. Students devise and use analytical schemes based on their own test results.

This approach teaches a great deal of chemistry and keeps student interest high.

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Laboratory Heating Bath Using Fluidized Solids

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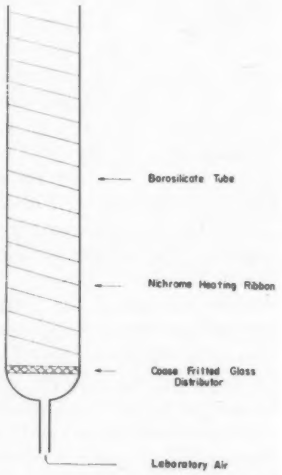
The laboratory heating bath described herein combines the high temperature limit of the traditional sand bath with the good heat transfer and temperature uniformity of fluidized beds.

The bath used in our laboratory (to heat a 2-ft. vertical reaction tube) consists of a nichrome-wrapped borosilicate glass tube, with a coarse fritted disc sealed near the bottom to contain the sand. Laboratory air enters the bottom inlet tube and passes through the fritted glass distributor, fluidizing the sand. For example, 20 × 40 mesh sand is fluidized in a 2-in. o.d. tube (about 30 in. in length) with air at about 20 l/min. Smoother operation was observed using 80 × 200 mesh silica gel. At 300°C, and an air flow rate of 6 l/min, a temperature variation of ±2°C was observed from a point 1 in. above the distributor plate to the top of a 10-in. bed. Further refinement in technique should probably give smaller temperature variation through the usable bed length. The highest temperature sought has been 455°C; undoubtedly one can get much higher temperatures with this sort of system. There was some tendency for solids to blow out of the bed, which was corrected by a glass wool plug.

None of the specifications or limits given here are critical. The system is recommended for its simplicity and its easy attainment of high temperatures.



Laboratory heating bath using fluidized solids.

To the Editor:

I was very interested to read the note on Fehling's test by Daniels, Rush, and Bauer in your JOURNAL recently, **37,** 205 (1960). I have spent a great deal of time trying to convince students that this is not a specific test for aldehydes. Usually they do not believe me. When it is "in the book," the printed word carries great conviction.

I would point out, though, that Tollens reagent is *not* an "infallible test" for aldehydes as stated by Daniels, *et al.* Fructose is one ketone which certainly reacts with Tollens reagent, and I believe that all α-hydroxyketones do.

D. A. H. TAYLOR

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Chem Ed

BOOK REVIEWS

From Atoms to Stars

Theodore A. Ashford, St. Louis University, Henry Holt & Co., New York. 1960. ix + 645 pp. Figs. and tables. 19 × 25 cm. $8.50.

This is an excellent text, well designed to accomplish the purpose for which it was written: "to give the student an understanding of the physical world as conceived by the scientists of our time." The difficult points are clarified by consistent and regular use of familiar example. Thus, the conclusion that the Law of Multiple Proportions indicates that matter is composed of "packages" called atoms is compared with the problem of determining whether the guests at a tea party sweetened their tea with granular sugar or with sugar cubes.

Each chapter is composed of three to five sections. The first sections discuss the appropriate topics, indicating the state of current knowledge and theory. The remaining sections show why we hold these theories to be probable and extend the discussion further, either qualitatively or, often, quantitatively. However, the mathematics is never difficult; and when it is less than easy, the engaging style of the author will encourage the student to continue diligently. I am particularly impressed with the consistent presentation of a theory as a concept which depends upon facts (rather than the converse) and with the very adequate discussions of these facts.

After a brief introduction, this text considers astronomy, then mechanics, electricity, light, and the structure of matter, each in separate chapters. The next six chapters discuss chemistry, molecules, the kinetic theory, reactions, the Periodic Table, atomic structure, and the relation between atomic structure and chemical properties. A chapter on nuclear energy follows; and the text concludes with two broad chapters, one on the stellar universe, the other on geology.

Each chapter concludes with a list of Notes and Exercises; many of the exercises are stimulating, few require mere recall of textual material. An appendix of 13 pages summarizes mathematical techniques, from simple multiplication through exponents to trigonometry. The index is complete and detailed. The illustrations are numerous (more than one per page), either specifically designed for this text or carefully selected from other sources. Every illustration adds to the discussion in the text.

An annotated bibliography of other books and of articles from the *Scientific American* is organized by chapters. Where appropriate, these selected references carry the discussion further than is possible in the text, and specific indications of the proper extended references are made at these places in the text.

I usually give my review copies to our librarian. This time, I will retain possession and instead suggest that several copies be purchased. We offer no course that can use this text, but our students will be encouraged to refer to it nevertheless. I recommend it highly, both for this use and as the text in a physical science course for students whose primary interests lie in other directions.

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Quantum Chemistry: Methods and Applications

R. Daudel, R. Lefebvre, and *C. Moser*, Centre de Mecanique Ondulatoire Appliquee du Centre National de la Recherche Scientifique, Paris, France. Interscience Publishers, Inc., New York, 1959. xiii + 572 pp. Figs. and tables. 16 × 23.5 cm. $14.50.

The scope of this book is very accurately described by its title: it deals thoroughly with the *procedures* of quantum mechanics rather than its principles; herein lies its very major achievements as well as its drawbacks.

The book is quite unique in discussing in detail the various conceptual and calculational techniques by which quantum mechanics is applied to problems in chemistry. By showing the details involved in the application, it goes far beyond the rather sketchy discussion of the principles of the approximation methods involved that are to be found in other books. It clearly demonstrates how quantum mechanical methods can be used to calculate, correlate, or understand a large variety of phenomena of chemical interest.

The book is divided into two parts: Part I. Empirical Methods, occupying somewhat more than half the book, makes only the most limited demands on the reader's mathematical training or ability. After a very cursory glance at generally important results of quantum theory and principles of wave mechanics it discusses several variants of the LCAO-MO method and the valence bond method in great detail. Separate, successive chapters discuss the application of these methods. The discussion throughout is meticulously clear and easy to follow.

Part II, Nonempirical and Semiempirical Methods, requires considerably more mathematical sophistication. It discusses from a more fundamental and critical point of view the nature of the empirical methods taken up in Part I. It also delves further into general features of the problems and fundamental techniques. After a thorough discussion of one electron systems, the helium atom and the hydrogen molecule, the orbital approximation as such for the treatment of polyelectronic systems is taken up in detail. The next chapter shows the application of the various approaches presented previously to the detailed study of two examples, ethylene and benzene. The last chapter discusses some recent developments, especially those along the lines of the so-called semiempirical approach including those based on the Pariser and Parr work and Moffitt's "Atoms in Molecules" approach. A set of three appendices deals with ancillary matters having higher mathematical content.

Reviewed in this Issue

Theodore A. Ashford, From Atoms to Stars

R. Daudel, R. Lefebvre, and *C. Moser*, Quantum Chemistry: Methods and Applications

Walter G. Berl, Editor, Physical Methods in Chemical Analysis. Volume 1, Second Revised Edition

H. J. Emeléus and J. S. Anderson, Modern Aspects of Inorganic Chemistry

Maurice Morton, Editor, Introduction to Rubber Technology

Cecil J. Schneer, The Search for Order

Garth L. Lee and *Harris O. Van Orden*, General Chemistry

H. Kalmus and *S. J. Hubbard*, The Chemical Senses in Health and Disease

Morris B. Jacobs, The Chemical Analysis of Air Pollutants

Georg Lockmann, The Story of Chemistry

Paul Pascal, Editor, Nouveau Traité de Chimie Minérale. Volume 7 (in two parts)

Proceedings of the International Symposium in Microchemistry

N. H. Hartshorne and *A. Stuart*, Crystals and the Polarizing Microscope

Alfred N. Martin, Physical Pharmacy

The absence of any treatment of the principles of quantum mechanics in the book is regrettable; an understanding of these is essential in order to realize that the importance of quantum mechanics far exceeds its use as a mere tool. Only such an understanding makes possible to view both the successes and failures of the applications in proper perspective. The danger of constructing imposing structures on most slender foundations can otherwise be very great. These strictures certainly apply only to the first part of the book, but it is there that the danger of "cook-book" application lies.

There are a few small flaws in the book, partially connected with this deemphasis of principles. Thus, for example, the implication that quantum mechanics deals with probabilities, "since no experiments have as yet been devised to determine the trajectories of electrons in atoms and molecules" (p. 11) is at least misleading. The probabilistic element in quantum mechanics is very much more fundamental than that.

The reminder in the book that much of chemical binding can be understood on the basis of a very simple and immediately comprehensible electrostatic picture (as, e.g., in the treatment of the H_2 and Li_2 and B_2H_6 molecules, bond angles, etc.), is most refreshing and commendable; however a clear exposition of the relation of this picture to the formalism of the LCAO or VB method is lacking. The book is well organized and clearly written; the type is excellent and the indexes good.

The flaws mentioned earlier are very minor and are vastly overshadowed by the outstanding features of the book as a whole. It is a first rate addition to the literature in an important field and, indeed, in its chosen area it is unique. Used in conjunction with any one of the many books which discuss the fundamental principles of quantum mechanics (on various levels of sophistication and mathematical complexity) it can be highly recommended both for self study and textbook use.

E. M. Loebl
Polytechnic Institute of Brooklyn
Brooklyn, New York

Physical Methods in Chemical Analysis Volume 1, Second Revised Edition

Edited by *Walter G. Berl*, Applied Physics Laboratory, John Hopkins University, Silver Spring, Maryland. 2nd revised ed. Academic Press, Inc., New York, 1960. xiv + 686 pp. Figs. and tables. 16 × 23.5 cm. $19.

This two volume set appeared in 1950–51 and the second edition of Volume I continues the aims of the earlier edition (J. Chem. Educ., **27**, 534 (1950); *Ibid.*, **28**, 399 (1951)); namely, a description of "those physical methods that have either proved themselves of considerable value in quantitative analytical work or are destined to play an important role in the future."

The contents include: Absorption Phenomena of X-Rays and γ-Rays (25 pp.), by G. L. Clark; X-Ray Diffraction Methods As Applied to Powder and Metals (100 pp.), by W. L. Davidson; X-Ray Diffraction as Applied to Fibers (53 pp.), by J. A. Howsmon and N. M. Walter; Spectrophotometry and Absorptimetry (63 pp.), by W. R. Brode and M. E. Corning; Emission Spectrography (77 pp.), by J. Sherman; Infrared Spectroscopy (61 pp.), by H. H. Nielsen and R. A. Oetjen; Raman Spectra (21 pp.), by J. H. Hibben; Refractive Index Measurement (50 pp.), by L. W. Tilton and J. K. Taylor; Mass Spectrometry (82 pp.), by C. F. Robinson; Electron Microscopy (70 pp.), by R. D. Heidenreich and C. J. Calbrick; Electron Diffraction (41 pp.), by W. C. Bigelow.

The coverage of the various topics is not comparable to the current importance of such topics in the literature of analytical chemistry; yet, each chapter is well complimented by adequate graphs, figures, charts, and references. As was the earlier edition, this volume will be of interest to teachers and industrial analytical chemists as a single volume reference book, with the realization that the scope of the text does not permit a complete or exhaustive treatment of any of the topics.

James M. Pappenhagen
Kenyon College
Gambier, Ohio

Modern Aspects of Inorganic Chemistry

H. J. Emeléus, University of Cambridge, and *J. S. Anderson*, Director, National Chemical Laboratory, Great Britain. 3rd ed. D. Van Nostrand Co., Inc., Princeton, New Jersey, 1960. xi + 611 pp. Figs. and tables. 14.5 × 22 cm. $7.75.

This revision of a well-known text is an essential purchase for those who do not own the previous (1952) edition. Although lengthened by only 54 pages, it contains a fair amount of new material, thanks to judicious paring, which has preserved virtually all the original contents with the exception of one chapter, Free Radicals of Short Life. The extent of revision is decidedly uneven, ranging from entirely new chapters to those in which the only changes are renumbering of figure- and reparagraphing. For those who already own the second edition, a coms parison of the two volumes may be useful.

The treatment of coordination compounds has been expanded to four chapters. The first two, Werner Theory and Inorganic Stereochemistry and Constitution and Valency Problems, present an excellent, over-all view of the field, both classical and contemporary, as well as thorough discussion of the newer theories of the coordinate linkage, including π-bonding and ligand field theory. The third and almost completely new chapter, Stability and Characteristic Reactions of the Co-ordinate Complex, with its emphasis on kinetics and reaction mechanisms, reflects the increasing use of physicochemical methods in inorganic chemistry. The fourth chapter, Metal Carbonyls and Other π-Bonded Complexes, twice the length of its counterpart in the previous edition, abounds in new information on syntheses, structures, and reaction mechanisms of metal carbonyls and nitrosyls. Several sections are devoted to electrophilic and nucleophilic attacks, concepts not even mentioned in the last edition. Entirely new sections on metal derivatives of saturated and unsaturated hydrocarbons contain material dating from the recent (1951) discovery of ferrocene.

The chapter Some Interstitial and Non-Stoichiometric Compounds has been considerably expanded to take full account of new experimental and theoretical studies of refractory carbides, nitrides, and borides. Many graphitic compounds are discussed in terms of molecular orbitals. In Recent Chemistry of the Non-Metals, the sections on sulfur nitrides, phosphonitrilic halides, interhalogen compounds, and polyhalide anions have been updated. The chapter Reactions in Non-Aqueous Solvent Systems is now divided into protonic and non-protonic solvents. In view of recent interest in bromine trifluoride and metal-ammonia solutions, the omission of these topics might be questioned.

The chapter on Valency presents more detailed treatments of electrovalency, electronegativity, and the Born-Haber cycle, and several sections, notably the one on resonance, have been rewritten from a molecular orbital viewpoint. Hydrogen and the Hydrides contains new material on boron hydrides and borohydrides as well as a very necessary discussion of salt-like hydrides, which had been lacking in the second edition. Radioactivity and Atomic Disintegration and Recent Chemistry of the Radioactive Elements now include information on transuranic elements through number 102.

Six of the 17 chapters remain essentially the same: Atomic Structure and the Periodic System; Atomic Weights and Isotopes; The Constitution of Inorganic Compounds (still inadequate in its discussion of hydrogen bonding); Poly-Acids and Silicates; The Peroxides and Per-Acids; and Metals and Intermetallic Compounds.

The book contains many useful graphs, tables, and diagrams. Some of the references are as recent as 1959. The topical arrangement makes individual chapters suitable for student reading assignments.

George B. Kauffman
Fresno State College
Fresno, California

Introduction to Rubber Technology

Edited by *Maurice Morton*, The University of Akron, Akron, Ohio. Reinhold Publishing Corp., New York, 1959. v + 547 pp. Many figs. and tables. 16 × 23.5 cm. $10.

This book represents a compilation of selected lectures on rubber technology, designed to introduce new members of the rubber industry to the basic aspects of this field. The lectures presented under twenty different chapter headings, were selected from various courses on rubber technology sponsored by local rubber groups in all parts of the country. In most cases, the authors had participated in these courses as instructors.

(*Continued on page A108*)

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by William L. Jolly
University of California, Berkeley

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1960 224 pp. Text price: $6.00

Organic Chemistry

by Keith M. Seymour, Butler University

1961 approx. 352 pp. Text price: $6.75

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by Alan F. Clifford, Purdue University

1961 approx. 576 pp. Text price: $6.95

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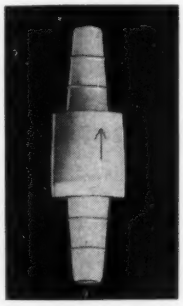

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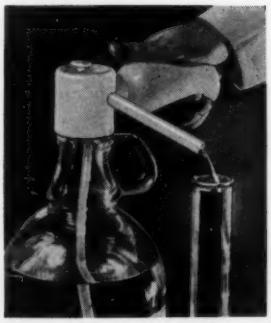

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BOOK REVIEWS

In general, the twenty subjects covered include the most important aspects of the field of rubber technology. A brief history with statistics to show the growth and development of rubber technology over the years makes interesting reading for scientists in other areas as well as being essential background for the rubber technologists. An introduction to polymer chemistry is included, since the rubber industry has found itself more and more involved in synthetic rubbers. Descriptions of the various polymeric materials of particular importance in the rubber industry are also included. While more adequate descriptions of these polymers are available from specialized sources, the material presented in the book is adequate for the purpose. The book is not intended to be a theoretical treatise and therefore does not include an elaborate discussion of modern theories of valcanization, a thorough treatment of the stereochemical aspects of polymeric materials, nor in many cases, a summary of the chemical changes which occur in certain of the processes. Compounding, physical testing, and reclaimed rubber are discussed. A chapter on processing equipment including some interesting historical background on this phase of the industry is also included.

The format is good, the tables, although reproduced by an offset process, are well done, and the book is relatively free of typographical errors. In general, most chapters are well documented; however, in certain instances, complete documentation would have been an insurmountable task due to the voluminous amount of material available on the subject.

As is to be expected, the writing style of the 25 contributors varies. For this reason, and because repetition and duplication is difficult to avoid in an endeavor of this type, the editor is to be commended in selection of material which avoids excessive repetition and results in a well integrated summary of the aspects of rubber technology.

GEORGE B. BUTLER
University of Florida
Gainesville

The Search for Order

Cecil J. Schneer, University of New Hampshire, Durham. Harper & Brothers, New York, 1960. xvii + 398 pp. Figs. 15 × 22 cm. $6. ($4.50 text edition.)

The necessity for opening lines of communication between scientists and the general public has been recognized as acute in recent years. Many books and many courses have been created to meet this necessity employing a wide variety of approaches to the problem. This book is a brilliantly conceived and executed attempt to present to the layman the spirit of science. Its historical and philosophical insights are quite sophisticated as is the general level of treatment

(*Continued on page A110*)

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S. Young Tyree, University of North Carolina, and Kerro Knox, Bell Telephone Laboratories

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1959, 851 pages, Ill., $7.95

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1952, 444 pages, Ill., $6.00

Coming Spring 1961

A SYSTEMATIC LABORATORY COURSE IN GENERAL CHEMISTRY, Second Edition

Harry H. Sisler; Jay J. Stewart, E. I. du Pont de Nemours & Co.; and William T. Lippincott, University of Florida

The Macmillan Company 60 Fifth Avenue, New York 11, N. Y.

of the subject matter considered. The author has thus chosen to deal with the problems of cosmology, mechanism, evolution, field theory, relativity and the quantum mechanics. And in each case he succeeds in developing with force and vigor a perspective, the present status, and the probable future impact of the problem on the development of both science and society.

The stories of the growth of the Newtonian world machine, organic evolution, and nineteenth century optimism are stimulatingly told. The role of mathematics is skillfully interwoven into the story. The dilemmas of modern physics are presented with clarity.

But, in the reviewer's opinion, it is this very success which makes the book unsuitable for the layman or scientifically unsophisticated reader looking for the answer to the question—"What is science all about?" The essentially historical or chronological development helps to alleviate this shortcoming but does not solve it. It seems unlikely, for example, that the general reader will be able to cope with the development of the ideas of Faraday and Maxwell concerning the electromagnetic field if he encounters them here for the first time.

This is *not* to say that the conventional "gee-whiz" or "pills and plastics" approach so often encountered in science writing for the layman is to be preferred. But rather that a certain minimum of familiarity with the details of science is necessary before its philosophical implications can be appreciated. Perhaps it must be conceded that understanding cannot be imparted in one book or one course no matter how clearly written or presented.

However, there is no question but that the book is ideally suited to the scientifically literate reader who wishes to fit his specialized knowledge into the broad historical outlines of the subject or who wishes to gain further insight into the philosophical implications of scientific endeavor. As Professor H. Margenau writes in his laudatory foreword, "I developed a strong desire to see this sort of treatment in the hands of my physics and philosophy students." It would be especially suited to the beginning graduate student or senior science major who all too frequently has had no time in which to relate the subject matter of many individual courses to the discipline as a whole.

RUSSELL H. JOHNSEN
Florida State University
Tallahassee

General Chemistry

Garth L. Lee and *Harris O. Van Orden*, both of Utah State University, Logan. W. B. Saunders Co., Philadelphia, Pennsylvania, 1960. vi + 637 pp. Figs. and tables. 16.5 × 24 cm. $7.25.

Although not specifically so stated in the preface, this book appears to be intended for students who lack previous preparation in chemistry. The subject matter falls into three divisions: chapters 1 to 7 covering general definitions and introducing atomic structure and bonding, chapters 8 to 26 containing a more extensive description of the elements with particular emphasis on their periodic relationships interspersed with chapters dealing with principles and theories, and chapters 27 to 37 devoted to organic chemistry. The amount of space allotted to organic chemistry (30%) is somewhat greater than is usual in a general chemistry textbook.

The textual material is easy reading and the illustrations profuse and clear. The diagram showing the operation of the mass spectrograph (p. 27) is particularly good. A few of the illustrations, however, (e.g., 8.7 p. 91) do not add much while others (e.g., 17.5 p. 251 with accompanying description) might better be used in a laboratory manual. Care is taken in working out problems so that the student can follow each step with ease. The subject of logarithms is appropriately introduced in an elementary and understandable manner.

In a few places terms are used which are not in accord with common usage. These include "decimal system" for metric system, "physical reactions," "tanks" for gas cylinders, "hydrogen salt" instead of acid salt, and "lye water" for sodium hydroxide solution.

Some inaccurate, or at best, misleading statements were found; for example, "Bismuth . . . [forms] most of its compounds by loss of the five valence electrons." "Electron hurled from a radium atom." The definition of an atom (p. 42) would exclude ordinary hydrogen.

(*Continued on page A112*)

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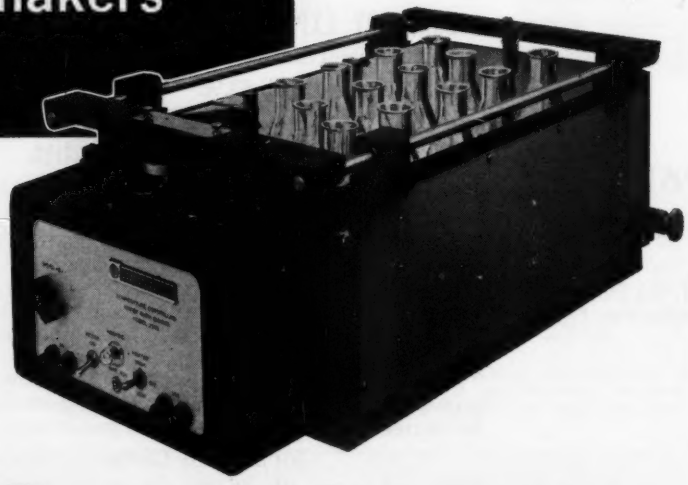

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1961. Approx. 776 pages. Prob. $8.75.

Kinetics and Mechanism

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Second Edition

By ARTHUR A. FROST *and* RALPH G. PEARSON, *Northwestern University.* The authors have thoroughly revised their book in order to provide both students and instructors with information which is completely up-to-date. They continue to emphasize the complexities of chemical reactions and the close relation of kinetics to mechanism.

1961. Approx. 432 pages. Prob. $10.00.

Organic Coating Technology

Volume II
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By HENRY FLEMING PAYNE, *University of Florida.* The phenomenon of color and its measurement is discussed together with the relation of hiding power of pigments to their physical-chemical characteristics. Specific formulations are offered in support of the basic theory of the physics and chemistry of pigments and coatings.

1961. 1363 pages. $17.50.

Electrode Processes

Transactions of the Symposium on Electrode Processes, Philadelphia, May 1959

Edited by ERNEST YEAGER, *Western Reserve University.* Includes all the papers presented at a symposium sponsored by the Electrochemical Society, and represents the first time in recent years that the work of so many internationally prominent electrochemists (from ten countries) has been presented in one publication.

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Viscoelastic Properties of Polymers

By JOHN D. FERRY, *University of Wisconsin.* In an exacting and uniform treatment of the viscoelastic properties and behavior of polymers, the author carefully develops a discussion of the phenomenological theory of viscoelasticity, followed by the presentation of a wide variety of experimental methods and a critical appraisal of their applicability to polymeric materials of different characteristics.

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The Chemistry of Terpenes

By A. R. PINDER, *University of Wales.* Contains the essentials of terpene chemistry and gives special regard to the reactions involved in degradation and synthesis, the interesting chemical transformations encountered in the field, and some of the more recent developments concerning stereochemistry and biosynthesis. Throughout the book verbal discussion has been reduced by the use of self-explanatory flow-sheets for many degradations and syntheses.

1961. 223 pages. $8.00

Microbial Cell Walls

By MILTON R. J. SALTON, *University of Manchester, England.* Describes the general physical, chemical and biochemical properties of microbial cell walls which constitute the major structural components of microorganisms. (One of the CIBA Lectures in Microbial Biochemistry.)

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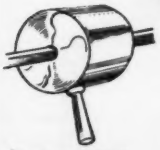

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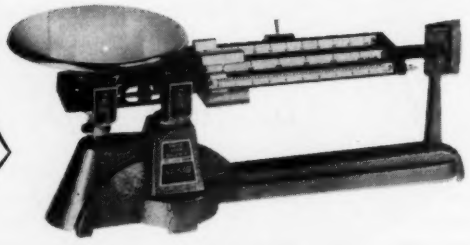

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BOOK REVIEWS

There is a generous supply of questions and problems at the ends of the chapters, although some of the questions seem a trifle unimaginative.

The treatment of ionization is well handled. In particular, an ingenious set of experiments is described to illustrate ionic equilibrium. A number of flow sheets are included which give a very clear picture of the processes under discussion. The principle of dipole moments is presented with clarity. The portion on organic chemistry is well handled, molecular models and reaction mechanisms being illustrated in a most understandable manner.

On the whole, this text should prove quite satisfactory for the elementary chemistry student.

CARL R. MELOY
University of Illinois
Chicago

The Chemical Senses in Health and Disease

H. Kalmus, University of London, and *S. J. Hubbard*, University College, London. Charles C Thomas, Publisher, Springfield, Illinois. vii + 95 pp. Figs. and tables. 15.5 × 23.5 cm. $3.75.

The authors of this little book have attempted to discuss all conceivable aspects of the senses of smell and taste, viz., the everyday uses of these senses and their survival value in evolutionary processes, the chemical nature of the stimuli, chemical and physiological theories of smelling and tasting, the anatomy and physiology of the sensory organs and nerves, genetic differences in perception, and the practical details of setting up tests for evaluating foods, cosmetics, repellents, etc.

Because of this broad coverage, different parts of the book will appeal more or less strongly to different sorts of readers. The chemist will appreciate the discussion of the complexity of the chemical stimuli, and will be interested but disappointed to learn that no generally accepted theory yet exists relating chemical structure to the sensation perceived. The intelligent layman, which includes the chemist when non-chemical topics are discussed, will probably enjoy the sections on the role of chemical senses in survival and adaptation, on the comparative sensory acuity of primitive and civilized peoples, on the psychological aspects of tasting and smelling, and on human genetic polymorphism with respect to tasting various substances. The physiological parts will be beyond the average chemist, but not the biochemist with a physiological background, and the discussions of how to set up tests to determine whether margarine really tastes different from butter, or which brand of chocolate is preferred by schoolboys, will be of minimal interest to anyone outside the food and advertising industries.

The more than 50 references appended should be sufficient to guide the reader in learning more (or indeed all there is to

(*Continued on page A114*)

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.....this month McGraw-Hill publishes the Second Edition of SIENKO and PLANE. The authors have altered the text only after thorough and careful examination. The changes have resulted in increased depth and lucidity—neither the scope nor the character of the book have been altered. Here's what the Journal of Chemical Education said about the first edition.....

"It is unusual in these days of many textbooks of introductory college chemistry to find one which is truly novel in more than one or two respects. 'Chemistry' is such a book. From the first glance at the attractive, almost-white cover, to a thorough perusal of the contents, one is impressed by numerous fine features. It is obvious that the writing and publishing were conceived and carried out with considerable thought and care. The use of black, white and gray for the many excellent drawings, diagrams, and tables gives unusual interest and clarity to the presentation. In addition, the textual material is printed in clear, easily read type, and organized exceptionally well for student study.

"Each chapter is headed by an appropriate illustration depicting the subject matter to follow. Principal sections within each chapter, as well as figures, tables, and exercises are numbered clearly for easy cross-reference purposes, and the chapter and section numbers are carried on each page for convenience. Each problem and exercise is labeled to suggest the principle or concept with which the problem deals.

"There are eight appendixes which present supplementary materials on nomenclature, mathematical operations, definitions from physics, the use of units in calculations, data on vapor pressure of water and oxidation potentials, and a few general references.

"The authors state in the preface that they 'have been guided by the observation that students respond to an intellectual challenge in proportion to the magnitude of the challenge.' With this thought in mind, they have gone 'deeply into the fundamentals of chemistry' and covered 'certain topics not normally discussed in elementary texts.' They state their belief that 'this approach does not make the subject more difficult but actually makes it clearer.' While this premise is undoubtedly valid, it is also most certainly true that different students have varying abilities and capacities for meeting intellectual challenges, and many a freshman will find Sienko and Plane's Chemistry a very rough hurdle to leap, or even to climb. This statement is not intended to detract from the excellence of the presentation, for it is, indeed, well done. However, it is a book for students of ability, and as such, it is one of the best books published to date.

"A number of recent 'modern' chemistry texts are organized similarly, with the first part devoted to theoretical principles and the latter part to descriptive chemistry. Many teachers object to this arrangement, feeling that abstract principles and theory tend to discourage the beginning student before he develops a confidence in the subject matter. The novelty of the presentation in this case is that so much of descriptive chemistry is woven skillfully into the theoretical material in the form of examples and illustrations that the objections to this arrangement are largely overcome. Much of the everyday chemistry of water, hydrogen, oxygen, the atmosphere, and other familiar materials is presented in this manner in the first half of the book.

"The authors confess that they include more materials than are usually found in elementary chemistry texts. Probably many teachers would choose to omit a few sections here and there. It is a rather firm conviction of this reviewer that the general introductory course of today attempts too much coverage with too little depth. In this book, the depth is quite adequate, and the breadth of coverage in terms of subject matter areas can be left to the discretion of the individual teacher.

"The problems and exercises are plentiful and well adapted. Types of calculations are presented in the text with numerous examples. The method of calculation falls just short of the so-called 'factor-label' method preferred by many. It would not be too difficult to go the rest of the way by setting up the final expressions for solving, with units, in a single numerical expression. Section 5 of Appendix 3 is suggestive of this procedure.

"An 'Instructors Manual' for the teacher accompanies the textbook. It includes a topical outline used by the authors for a one-year course, a list of lecture demonstrations, sample examinations and quizzes, a mathematical aptitutde test, a list of possible laboratory experiments, materials for special problem sessions, and answers to those problems for which answers are not given in the book.

"In summary, this is a very worthy addition to the field of general chemistry textbooks. It is a book which every high school and college teacher would find of value in his own library, and many college teachers would find it stimulating for class use."

GRANT W. SMITH

The Pennsylvania State University
University Park, Pennsylvania

POLYMERIC MATERIALS

By Charles C. Winding, Cornell University, and Gordon D. Hiatt, Eastman Kodak Company. Ready in March, 1961.

Provides a reference background from which the reader can understand the engineering principles and applications of polymeric materials. An introduction to polymer chemistry, molecular structure, and properties is followed by a treatment of fabricating methods and applications, all involving fundamentals common to a large number of different polymers. This material is followed by a detailed discussion of specific commercial polymers.

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By F. J. C. Rossotti and Hazel S. Rossotti, University of Edinburgh. *McGraw-Hill Series in Advanced Chemistry.* Ready in March, 1961.

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THERMODYNAMICS, Second Edition

By G. N. Lewis and M. Randall (Revised by Kenneth S. Pitzer and Leo Brewer, *both of the University of California, Berkeley*) *McGraw-Hill Series in Advanced Chemistry.* Ready in February 1961

A revision by two top men in their field of a 1923 classic. The informal and much-admired style of the first edition has been retained and will make the book very appealing to all its readers. Because the last edition of the book was 37 years ago, there is much new material included in the new edition, particularly in the middle and late chapters. These chapters have now been brought up to the present frontiers of thermodynamics. Illustrative materials have been brought up to date. As before, illustrative problems are worked out in considerable detail.

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learn) about any specific topic which intrigues him.

In summary, if you do not know anything about the chemical senses, but would like to find out something about them, the 80 pages of this book could be perused with profit in about an hour, if the less appealing topics are omitted. Chapter 6, on diagnostic uses of smell, and on the changes in chemical sensory ability in disease, is hardly worth reading, and Chapter 7, the summary, is more confusing than helpful.

PETER OESPER
Hahnemann Medical College
Philadelphia, Pennsylvania

The Chemical Analysis of Air Pollutants

Morris B. Jacobs, Air Pollution Control Consultant, New York. Volume 10 of the Chemical Analysis Series. Interscience Publishers, Inc., New York, 1960. xviii + 430 pp. Figs. and tables. 15.5 × 23.5 cm. $13.50.

This book is Volume 10 of a "Series of Monographs on Analytical Chemistry and Its Applications" under the editorship of P. J. Elving and I. M. Kolthoff. Dr. Jacobs is also the author of Volume 1 of this series, "The Analytical Chemistry of Industrial Poisons, Hazards, and Solvents."

With the realization that air pollution is of increasing concern to civilization, the author has attempted to cover the complete scope of the topic, including sampling, equipment, analysis, and interpretation of results. The book is composed of 17 chapters, an appendix of 14 tables, and an adequate subject index. There are tables and figures throughout the text. Representative chapters include: Sampling, Sootfall and Dustfall Analysis, Suspended Particulate Matter, Sulfur-Bearing Air Pollutants, Organic Compounds, and Radiochemical Determinations.

The author's widespread knowledge of the problems of air pollution has produced a well organized text. It is unfortunate that the analytical chemistry portions of the book were not always well prepared. There are a number of inconsistencies in terminology. Thus, one finds the indiscriminate use of optical density, absorbance, transmission, and percentage transmittance. The reviewer objects to an analytical method "geared" to a particular instrument. For example, on page 44, is found "mgNH_3 = Klett reading × factor . . . . . ." The proposals range from the use of a Cary Model 11 recording spectrophotometer to the use of Nessler tubes. Such statements as "compare the color" and "read in spectrophotometer" may be both confusing and misleading. The trace and classical methods of analysis employed are, however, generally up-to-date and can be performed by a competent analyst. The passage devoted to vapor phase chromatography and infrared techniques for organic analyses is very incomplete and insufficient.

The book is the first in its field, and, for this reason, will be of value to all concerned

(*Continued on page A116*)

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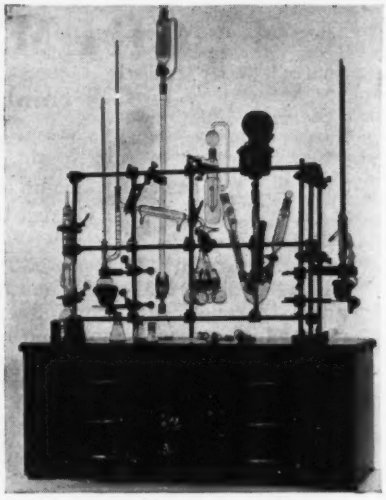

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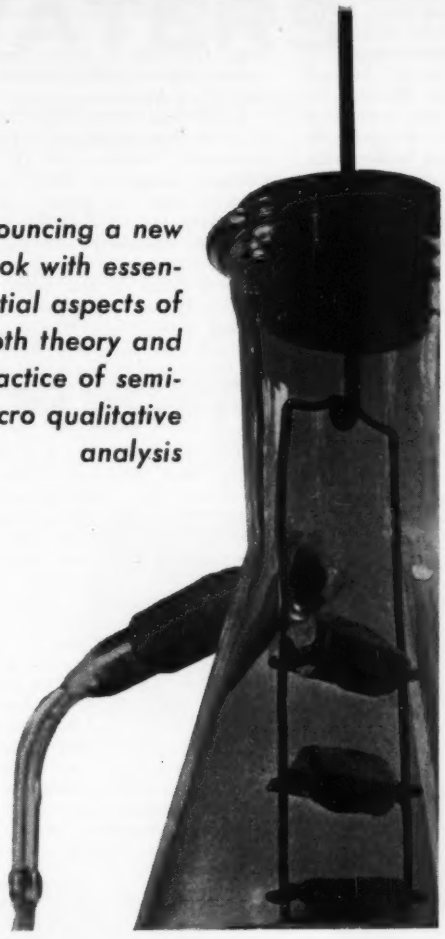

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JAMES M. PAPPENHAGEN
Kenyon College
Gambier, Ohio

The Story of Chemistry

Georg Lockemann. Philosophical Library, Inc., New York, 1959. 277 pp. 12 × 19 cm. $4.75.

Reading this book is a pleasure. Lockemann writes in a sprightly manner, not often the case with historians of chemistry, and he has a sense of humor that enlivens the work. His pleasant easy-going style is illustrated by the following passage: "[Priestley] was a fanatic of religious free-thought who could tolerate no ideas besides his own. Since he was also a brave and doughty fighter he could not remain for long at peace with his surroundings. . . . Once when he managed to stay beyond that term apparently fixed for him by nature, a raging mob set fire to the roof over his head and he barely escaped with his life. . . . Finally, having broken with one and all, he emigrated to America in 1794. He refused the offer of a professorship at the University of Philadelphia and spent ten years as a farmer in the area of the sources of the Susquehanna. There he died at 71, apparently the victim of a poisoning; but it is by no means certain that his wild life ended that way."

Lockemann's statement about Priestley being poisoned is one of the oddest fables in chemistry and one that has been popping up in European histories for more than a century. It is a mystery who started this myth, and it is remarkable that an otherwise careful historian would be snared by this old tale, particularly when there are so many carefully documented papers available concerning Priestley's life in America.

Lockemann writes from the viewpoint of a German historian. That is, he is thoroughly familiar with the history of German chemistry. He is perhaps less familiar with the history of European chemistry, but he is hardly familiar with American chemistry. He mentions only 25 Americans out of the more than 1600 persons listed in the index. He says practically nothing about chemical industry in the United States, although information on this subject has been available for years in Williams Haynes' 6 volume "American Chemical Industry." As a result the book appears to be a bit lopsided.

The physical make-up of the book requires some comment. First, there are no illustrations. This will not weigh unduly with a reader who is already familiar with history of chemistry, but it may dull the volume for someone who is using the book as an introduction to history. Secondly, it has no references. This may or may not be a drawback, depending on whether the reader is reading for pleasure or for information, and whether the reader has access to other works. Finally, it has no subject index. Again, this may or may not be an inconvenience, depending on the purpose of the reader.

This book would not be suitable for a text, but it would certainly be interesting collateral reading for a series of lectures on history of chemistry. Any chemist will enjoy it as light reading.

WYNDHAM D. MILES
The National Archives
Washington, D. C.

Nouveau Traité de Chimie Minérale. Volume 7 (in two parts)

Edited by *Paul Pascal,* Sorbonne. Masson et Cie, Paris, 1959. Two volumes totalling xxxix + 1474 pp. 551 figures, 363 tables. 17.5 × 26 cm. 200 NF.

The division of Volume 7 of Pascal's new treatise into two separate parts appears to have been done more for the convenience of binding than for logical separation of material. Paging is continuous, and a single index appears at the end of part two of the set. The volume is concerned with scandium, yttrium, the rare earth elements, and actinium, but scandium and yttrium are not considered separately from the lanthanides here, so that all but the last 60 pages may be considered to be a treatise on the rare earths. All material, except that on actinium by G. Bouissieres, was written jointly by Trombe, Loriers, Gaume-Mahn, and La Blanchetais.

These authors have produced an excellent piece of work on the rare earths, particularly on mineral sources, general methods of separation, chemical and physical properties, and separation by ion exchange. The treatment on metal-

(*Continued on page A118*)

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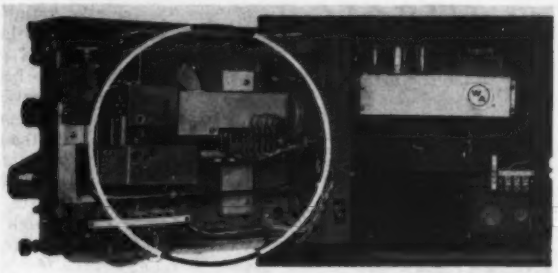

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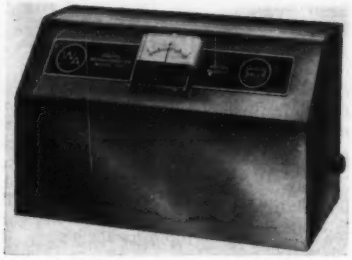

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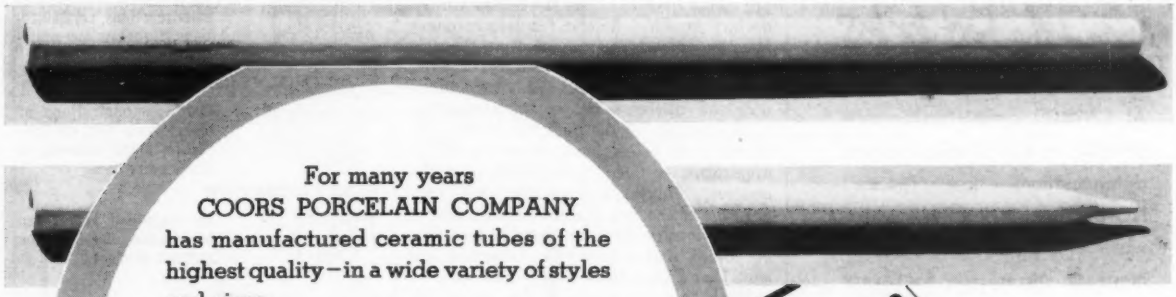

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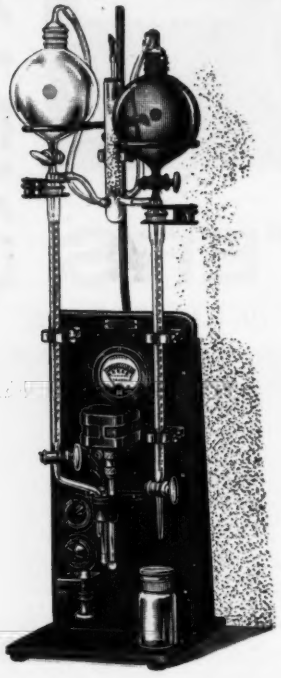

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lurgy is also quite good. The historical section presents considerable information in the space allotted.

The section on actinium consists of but 34 pages, and the literature search for it was conducted through 1959. Modern treatment has been given to its isotopes, nuclear properties, preparation, separation, and chemical properties. The literature searches for the rare earths were not conducted systematically beyond December 31, 1954, in most cases. Of the 56 bibliographic lists, only one went through 1958, five through 1957, and three through 1956, despite publication date of 1959. Despite a few such drawbacks, the volume is an excellent work and fills a need for a comprehensive reference book on the rare earths which uses modern concepts.

ROGER V. KRUMM
University of Florida
Gainesville

Proceedings of the International Symposium in Microchemistry

Published for The Society for Analytical Chemistry, Pergamon Press, Inc., New York, 1960. xxvi + 583 pp. Many figs. and tables. 18 × 25.5 cm. $15.

The Symposium on Microchemistry held at Birmingham University in August, 1958, was attended by over 400 analytical chemists including 110 from 25 countries outside the United Kingdom. Many of these participated in the extensive discussions which are reported after most of the 60 papers in these proceedings.

R. C. Chirnside's remarks on page 498 are worth quoting: "At the outset I must declare myself a heretic for I have never been completely convinced that there is such a thing as microchemistry or even microchemical analysis. To what indeed does the 'micro' refer; to the chemistry, to the sample, to the quantity of the substance to be determined, to the size of the apparatus, or even to the stature of the analyst? Having studied the whole programme of the Symposium I am no nearer a definition; indeed the range and variety of topics and techniques discussed are such that I am confirmed in my heresy."

With this concession in the use of the term "microchemistry," the reader will enjoy the authors' accounts of their research on nearly every analytical technique that can be considered "micro" in any sense. Included are evaluations, trends and forecasts, specific applications, descriptions of specific laboratories, and training. All of the papers are extremely well written—even the foreign papers have been cast in the style of the English authors. Major topics and authors are: Qualitative Micro-analysis, Feigl, Welcher, West, and Weisz; The Decomposition of Organic Matter, Diehl and Smith, and Pijck, *et al.*; Weighing, Hodsman and Corner; Classical Methods for the Determination of Elements, Groups and Physical Constants, Lieb, Schöniger, Malissa, Colson, Levy, Kirsten, Hersch, Ma, Veibel, Stephen, and Sobotko; Biochemical Methods, Adamson and Simpson, Nicholas, and Rosen; Chromatography, Crouch and Swainbank, Grand-Clement,

(*Continued on page A121*)

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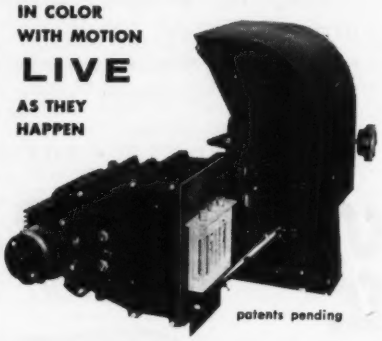

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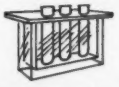

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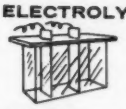

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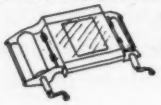

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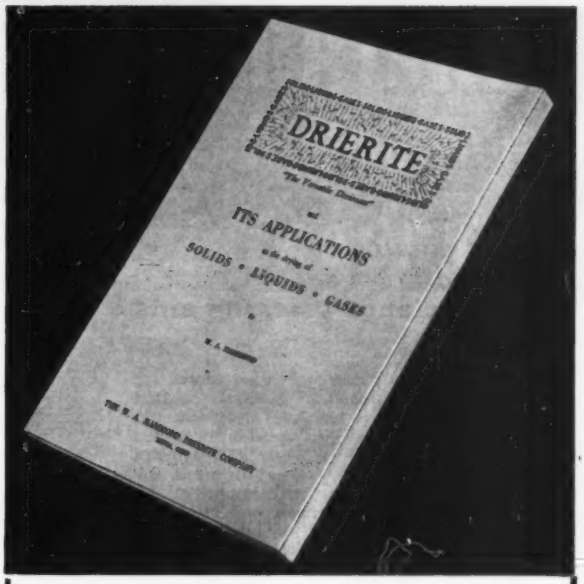

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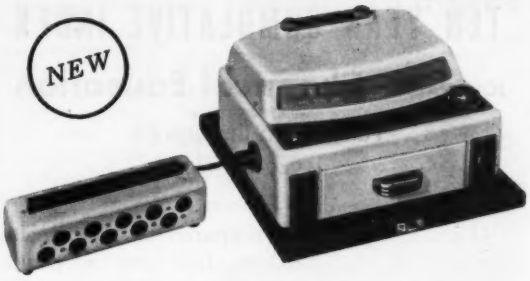

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It is difficult to single out individual papers for comment in the space available. The papers on methods and techniques will be valuable to all analytical chemists as concise summaries of current practise. Brief reviews of theoretical principles and key references are given.

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ROBERT L. PECSOK
University of California
Los Angeles

Crystals and the Polarizing Microscope

N. H. Hartshorne, University of Leeds, England, and *A. Stuart*, University of Exeter, England. 3rd ed. St. Martin's Press, Inc., New York, 1960. xv + 557 pp. Figs. and tables. 14.5 × 22 cm. $17.50.

This edition as well as the previous publication is primarily aimed at those persons desirous of knowledge of crystals, their growth, and study. It must become apparent to the serious chemist that much useful information can be gained about compounds by use of the polarizing microscope. In view of the diminishing number of trained and qualified optical microscopists this book makes a good attempt to explain to the reader the valuable but also rather difficult science of crystals, their transformation, and methods of study by the use of rather little known (to the non-microscopist) equipment.

The 13 chapters and 544 pages of this publication (12 chapters and 462 pages in the 2nd edition) contain in general most of the information present in the previous edition. There has been some rearrangement of topics to create another chapter and to present a more logical arrangement of information. Most of the contents of whole chapters remains essentially unchanged, but there have been worthwhile topics added such as the discussion on crystal growth mechanism. While this information has been known and spoken of for many years by a few persons in microscopy, far too few scientists recognize its importance. The authors present this subject in a concise and clear manner. Its addition helps to make a more complete presentation of crystals, different types of crystals, their description, and methods of study.

The chapter concerning hot and cold stages has been modernized. The methods of study of the effect of temperature changes on mounting media (liquids) and the specimen variation are shown to yield much useful information which cannot be obtained by use of only one temperature. Not only are differences in refractive indices obtainable but also phase changes, identification by mixed melting points, etc.

In general the real value of this edition as well as the second edition is in the clarity and directness of presentation. The information on the difficult science of examination of crystals and textile fibers by the polarizing microscope is so well given so as to be a microscopist's handbook for reference purposes. The universal stage and its use, for instance, are described so as to make its use relatively easy. Any college course in microscopy or chemical microscopy will be enriched by the frequent use of this book.

In summation it may be said that this third edition contains a few items more than the second edition. For those chemists who do not now own the earlier edition, it will be a good addition to your personal library.

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Physical Pharmacy

Alfred N. Martin, Purdue University School of Pharmacy, Lafayette, Indiana. Lea & Febiger, Philadelphia, 1960. 692 pp. Figs. and tables. 16 × 24 cm. $15.

This book combines the basic concepts of classical physical chemistry with applications to pharmacy and medicine. Methods used in measuring the physical-chemical constants of medicinal chemicals makes the text invaluable to the study of pharmacy. The basic physical chemistry concepts are presented in such a way that the student is able to apply them to problems involving standardization, stability and absorption of drugs and drug products. Numerous examples of such applications are to be found in the text.

Although the calculus is used in the derivation of several of the fundamental equations, it is not a prerequisite to using the book. The explanations given and the problems included require only freshman mathematics for their understanding and solution.

The text is well written on good paper and is well illustrated. Wide use of tables and line drawings lend to the ease of understanding. It is up-to-date in all respects.

Despite the fact that the text is written primarily for pharmacy students and manufacturing pharmacists, it serves also as an ideal text for use in the premedical program as well as in any beginning course in physical chemistry. The emphasis on drugs and problems involving drugs makes the text readable and interesting to most students of physical chemistry.

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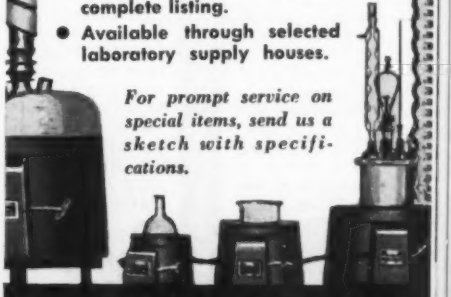

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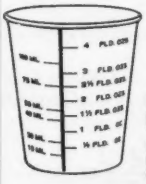

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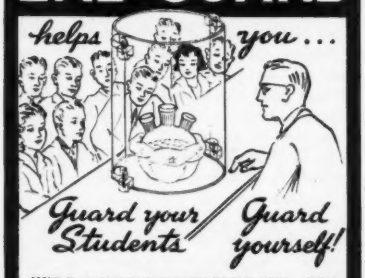

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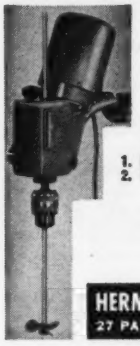

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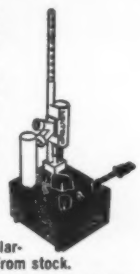

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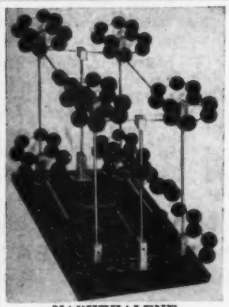

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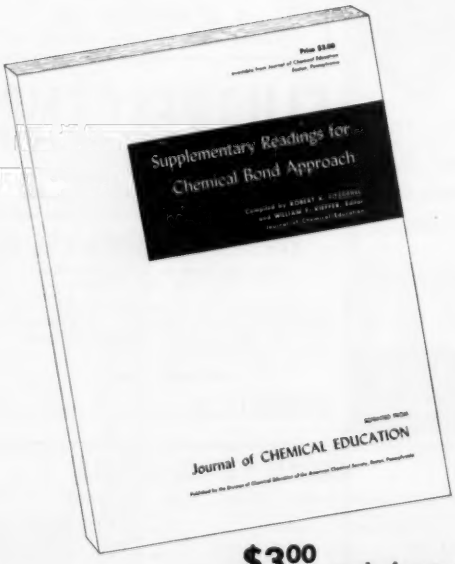

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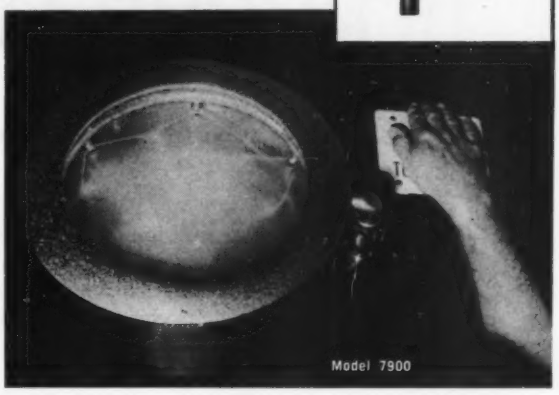

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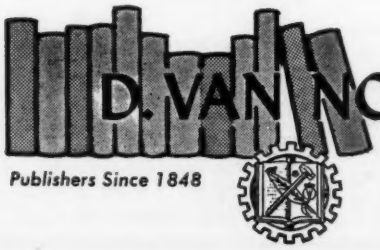

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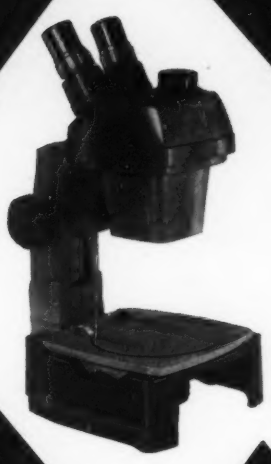

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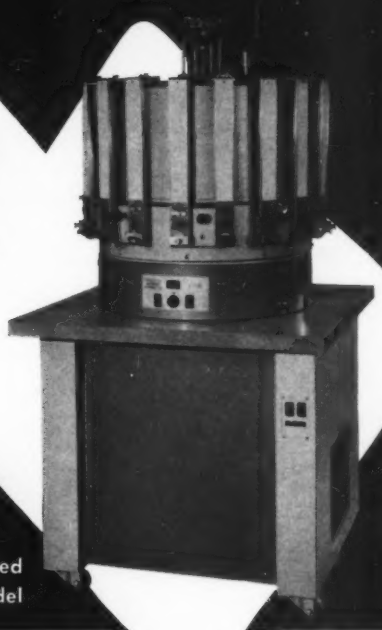

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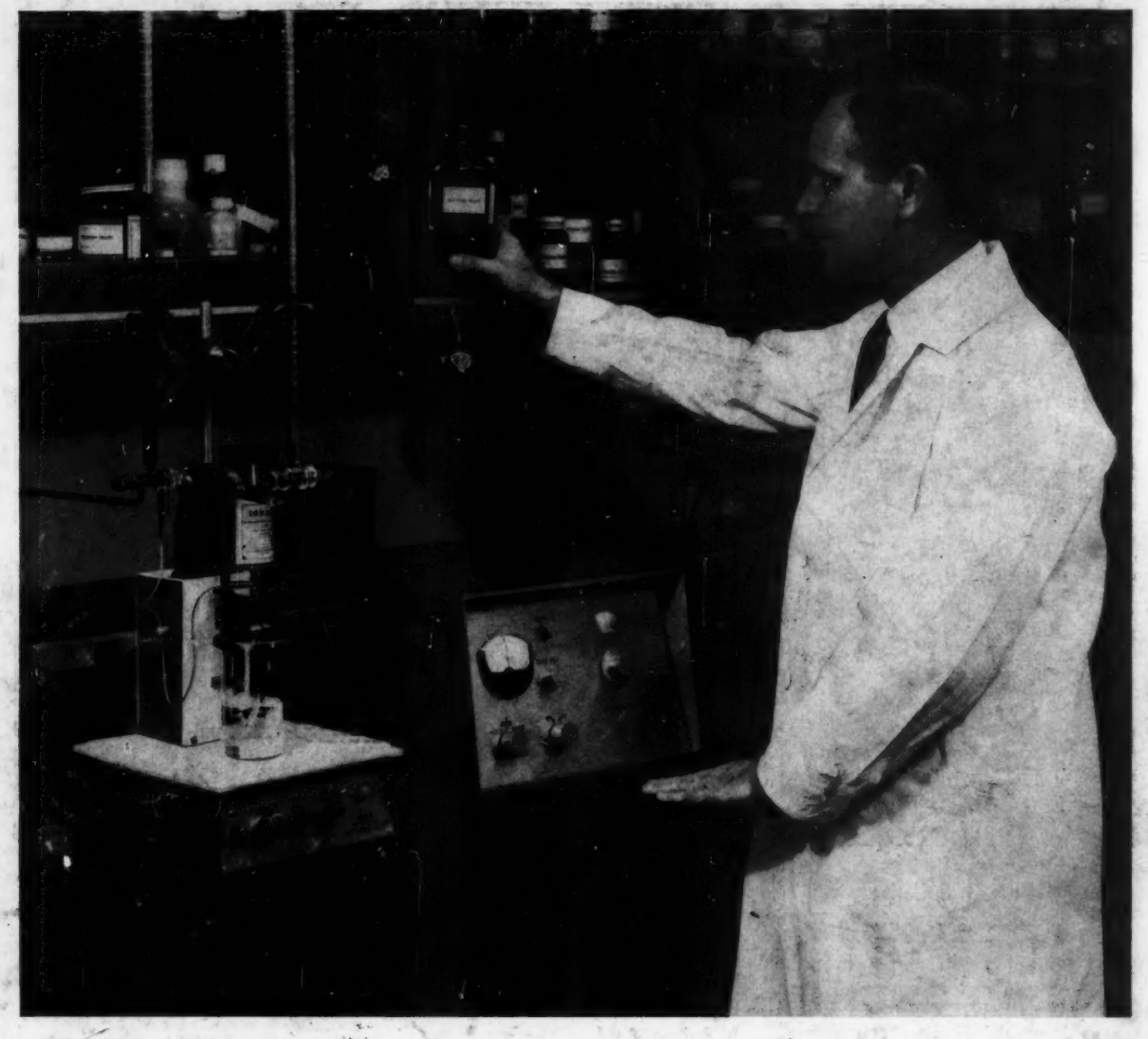

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